THE INTERACTION OF RADIATION AND MATTER: SEMICLASSICAL THEORY

I. REVIEW OF BASIC QUANTUM MECHANICS: CONCEPTS, POSTULATES AND NOTATION:

At the outset, let us, briefly, reconsider why quantum mechanics is necessary?

- Forces known in classical electrodynamics cannot account for the remarkable stability of atoms and molecules.

- Disturbed dynamic systems radiate only frequencies which may be expressed as differences between certain values (Ritz's Combination Law of Spectroscopy).[1]

- All physical systems -- *viz.* material "particles" and electromagnetic fields -- exhibit *wave-particle duality* -- *i.e.* to explain particular observations the system must in some instances be modeled as a particle and in others as a wave.

- There is a limit below which the disturbance associated with a observation is not negligible and, thus, there is an unavoidable *indeterminacy* in the prediction of observed results.

To proceed, we recall an ancient comment of P. A. M. Dirac:[2]

> "Quantum mechanics…requires the states of a dynamic system and the dynamical variables to be to be interconnected in quite strange ways that are unintelligible from the classical standpoint. The states and dynamic variables have to be represented by mathematical quantities of different natures from those ordinarily used in physics"

Following the Master we begin with the general quantum mechanical ***principle of superposition of states***. To quote him once more:

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[1] According to classical theory, a disturbed system should radiate certain fundamental frequencies and their harmonics. Each fundamental frequency would be associated with one of the systems degrees of freedom.

[2] P. A. M. Dirac, *The Principles of Quantum Mechanics* (Revised fourth edition), Oxford University Press (1967).

> "The non-classical nature of the superposition process is brought out clearly if we consider the superposition of two states, A and B, such that there exists an observation which, when made on the system in state A, is certain to lead to one particular result, **a** say, and when made on the system in state B is certain to lead to some different result, **b** say. What will be the result of the observation when made on the system in the superposed state? The answer is that the result will be sometimes **a** and sometimes **b**, according to a probability law depending on the relative weights of A and B in the superposition process. It will never be different from both **a** and **b**. *The intermediate character of the state formed by superposition thus expresses itself through the probability of a particular result for an observation being intermediate between the corresponding probabilities for the original states, not through the result itself being intermediate between the corresponding results for the original states.*
>
> "…The superposition process is a kind of additive process and implies that states can in some way be added to give new states. The states must therefore be connected with mathematical quantities of a kind which can be added together to give other quantities of the same kind. The most obvious of such quantities are vectors"

With this motivation, Dirac introduced the whimsical name ***ket*** and the ingenious right half-bracket notation -- $\boxed{|\ \rangle}$ -- to represent the vectors connected to the states of a quantum mechanical system.[3] Thus, the superposition of two states to form a third is represented in abstract ket vector notation, as

$$|R\rangle = c_1\,|A\rangle + c_2\,|B\rangle \qquad \text{[I-1a]}$$

where the c_n's may be complex numbers. More generally,

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[3] The state is specified **by the direction** of a ket vector and any length assigned to the vector is irrelevant.

$$|R\rangle = \sum c_\eta \, |\eta\rangle + \int c(\xi)\, |\xi\rangle d\xi \qquad \text{[I-1b]}$$

For any set of vectors one can always define a complementary set of dual vectors. In Dirac's development, the dual vectors are called ***bra*** vectors and denoted by the other half of the *bracket* symbol $\boxed{\langle\ |}$. A bra vector $\langle P|$ is completely defined when its scalar product -- *i.e.* $\langle P||Q\rangle \equiv \langle P|Q\rangle$ -- with every ket vector $|Q\rangle$ is known. The scalar product is linear (or anti-linear) in the sense that:

$$\langle B| \left\{ |A\rangle + |A'\rangle \right\} = \langle B|A\rangle + \langle B|A'\rangle \qquad \text{[I-2a]}$$

$$\langle B| \left\{ c|A\rangle \right\} = c\langle B|A\rangle \qquad \text{[I-2b]}$$

$$\left\{ \langle B| + \langle B'| \right\} |A\rangle = \langle B|A\rangle + \langle B'|A\rangle \qquad \text{[I-2c]}$$

$$\left\{ c\langle B| \right\} |A\rangle = c\langle B|A\rangle \qquad \text{[I-2d]}$$

$$\text{If } \langle P|A\rangle = 0 \text{ for all } |A\rangle, \text{ then } \quad \langle P| \equiv 0. \qquad \text{[I-2e]}$$

Although bras and kets are very different creatures, **it is assumed** that there is a one-to-one correspondence between bras and kets such that the bra corresponding to the ket $|A\rangle + |A'\rangle$ is the sum of the bras corresponding to the kets $|A\rangle$ and $|A'\rangle$ -- *viz.* $\langle A| + \langle A'|$ -- and the bra corresponding to the ket $c|A\rangle$ is the product of c times the bra corresponding to the ket $|A\rangle$ -- *viz.* $c\,\langle A|$.[4] It is also assumed (**an especially**

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[4] Dirac uses the words ***conjugate imaginary*** to connote a corresponding bra-ket pair and, thereby, draws attention to an important distinction. The words ***conjugate complex*** are reserved for the pairing of ordinary complex quantities which can be split into real and imaginary parts. In particular, the real part of a complex quantity is given by one half of the sum of the quantity and its conjugate complex. However, bras and kets are representations in dual vector spaces and cannot be added!

important assumption) that $\langle P|Q\rangle = \langle Q|P\rangle$ -- so that $\langle P|P\rangle$ is real -- and that $\langle P|P\rangle \geq 0$.[5]

To briefly recapitulate, the directions of vectors of a dual abstract vector space are to be associated with states of a dynamic system. To extend the Diracian view, dynamic variables and observables of the system are to be associated with (real) abstract **linear operators** which transform one vector into an other -- *viz*.

$$|F\rangle = Op\ |A\rangle\ . \qquad [\text{ I-3 }]$$

Linearity requires that $$Op\{|A\rangle + |A'\rangle\} = Op\ |A\rangle + Op\ |A'\rangle \qquad [\text{ I-4a }]$$

and $$Op\{c\,|A\rangle\} = c\ Op\ |A\rangle \qquad [\text{ I-4b }]$$

The conjugate imaginary or adjoint of $|F\rangle$ is obtained by operating (to the left) with the **hadjoint** of the linear operator [6] on the conjugate imaginary of $|A\rangle$ -- *viz*.

$$\langle F| = \left(|F\rangle\right)^{\dagger} = \left(Op\ |A\rangle\right)^{\dagger} = \langle A|\,Op^{\dagger}\ . \qquad [\text{ I-5 }]$$

By assumption $\langle P|Q\rangle = \langle Q|P\rangle$ so that

$$\langle A|\,Op^{\dagger}|F\rangle = \langle F|\,Op\ |A\rangle \qquad [\text{ I-6 }]$$

From which we may easily show that

$$Op^{\dagger\dagger} = Op \qquad [\text{ I-7a }]$$

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[5] We may thus identify the numbers $\langle P|Q\rangle$ as elements of a hermitian matrix -- Charles Hermite (1822-1901).

[6] If a linear operator corresponds to a dynamic variable, then the adjoint of that operator corresponds to the conjugate complex of the dynamic variable.

and
$$\left(Op_1\, Op_2\right)^{\dagger} = Op_2^{\dagger}\, Op_1^{\dagger} \qquad \text{[I-7b]}$$

To further develop the notion of linear operators, we examine the ***eigenvalue*** equation

$$\mathcal{A}\,|F\rangle = a\,|F\rangle \qquad \text{[I-8]}$$

where $\mathcal{A}$ is a linear operator and a is a number. In most instances, the equation arises in a context wherein $\mathcal{A}$ is a known linear operator, which corresponds to a dynamic variable, and the number a -- an eigenvalue -- and the ket $|F\rangle$ -- an eigenvector -- are unknowns to be determined. Only self-adjoint (*i.e.* real or Hermitian) linear operators are of any use quantum mechanics so we may write quite generally

$$\mathcal{A}\,|P\rangle = p\,|P\rangle \qquad \text{[I-9a]}$$

$$\langle Q|\,\mathcal{A} = q\,\langle Q| \qquad \text{[I-9b]}$$

It follows that all eigenvalues are real numbers, that eigenvalues associated with eigenkets are the same as those associated with eigenbras, and that conjugate imaginary of any eigenket is an eigenbra belonging to the same eigenvalue (and conversely). If we may assume that a complete set of eigenvalues and eigenvectors of a given dynamic variable ξ is known, we may use the following exquisitely self-explanatory notation to delineate the manifold of all eigenvalue equations:

$$\xi\,|\xi'\rangle = \xi'\,|\xi'\rangle\,; \quad \xi\,|\xi''\rangle = \xi''\,|\xi''\rangle\,; \quad \xi\,|\xi'''\rangle = \xi'''\,|\xi'''\rangle\,; \ldots\ldots . \qquad \text{[I-10]}$$

It is easy to show that **within this manifold, the eigenvectors belonging to different eigenvalues are orthogonal!**

REPRESENTATIONS AND QUANTUM CONDITIONS:

GENERAL NOTIONS:

As pointed out earlier, a bra vector is completely defined in terms of a set of numbers -- the ***representatives*** -- which are the values of the scalar product of the bra with every ket in a ***representation*** (and conversely). Thus, if we have a complete set of discrete, orthonormal state vectors $|\eta\rangle$ we may write a superposition of discrete states as

$$|\xi\rangle = \sum_{\eta} |\eta\rangle\langle\eta|\xi\rangle \qquad \text{[I-11a]}$$

If we have a complete set of continuous, orthonormal state ket vectors $|\eta\rangle$ we may write a superposition of continuous states as

$$|\xi\rangle = \int d\xi\, |\eta\rangle\langle\eta|\xi\rangle \qquad \text{[I-11b]}$$

Consideration of these two expansions leads to extremely valuable expressions for expansions of the **identity operator** -- *viz*.

$$I = \sum_{\eta} |\eta\rangle\langle\eta| \qquad \text{[I-12a]}$$

or

$$I = \int d\xi\, |\xi\rangle\langle\xi| \qquad \text{[I-12b]}$$

This is, essentially, a diagonal **dyadic** representation of the identity operator. More generally, a linear operator has the following dyadic representation

$$Op = \sum_{\xi''}\sum_{\xi'} |\xi'\rangle\langle\xi''|\langle\xi'|Op\,|\xi''\rangle \qquad \text{[I-13a]}$$

or

$$Op = \iint |\xi'\rangle\langle\xi''|\langle\xi'|Op\,|\xi''\rangle\, d\xi'\, d\xi'' \qquad \text{[I-13b]}$$

Since commuting linear operators have simultaneous eigenstates, it is a critical matter to know whether or not two observables commute and, if they do not, to know the *commutation relation* needed to replace the classical *commutative law of multiplication*. Fortunately, the so called method of ***classical analogy*** provides a systematic means for establishing such relationships for the very large class of dynamic systems which obey the Euler-Lagrange equations of motion.[7] If such a system has *n* degrees of freedom, it is describable in terms of n pairs of ***canonically conjugate*** coordinates q_r and momenta p_r.[8] The essence of the method of classical analogy is embedded in statement that the **commutator is the quantum mechanical equivalent of the classical Poisson Bracket** and is given by

$$[\mathrm{u},\mathrm{v}] \equiv \mathrm{uv} - \mathrm{vu} = i\hbar\{\mathrm{u},\mathrm{v}\}_{\mathrm{PB}} \equiv i\hbar\sum_r \left\{ \frac{\partial \mathrm{u}}{\partial q_r}\frac{\partial \mathrm{v}}{\partial p_r} - \frac{\partial \mathrm{u}}{\partial p_r}\frac{\partial \mathrm{v}}{\partial q_r} \right\} . \qquad [\text{ I-14 }]$$

Thus, the **fundamental quantum conditions** or **commutation relationships** are

$$[q_r, q_s] \equiv q_r\, q_s - q_s\, q_r = 0 \qquad [\text{ I-15a }]$$

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[7] This class includes fields, in particular the electromagnetic field, as well as the mechanics of particle motion.

[8] Precisely: A pair of dynamic variables r and s are said to be canonically conjugate if there is function of the two variables $\mathcal{H}(r, s)$ such that

$$\frac{dr}{dt} = \frac{\partial \mathcal{H}}{\partial s} \text{ and } \frac{ds}{dt} = -\frac{\partial \mathcal{H}}{\partial r} .$$

For a second function of the canonical variables $\mathcal{F}(r, s)$ we have

$$\frac{d\mathcal{F}}{dt} = \frac{\partial \mathcal{F}}{\partial r}\frac{dr}{dt} + \frac{\partial \mathcal{F}}{\partial s}\frac{ds}{dt} = \frac{\partial \mathcal{F}}{\partial r}\frac{\partial \mathcal{H}}{\partial s} - \frac{\partial \mathcal{F}}{\partial s}\frac{\partial \mathcal{H}}{\partial r} = \{\mathcal{F}, \mathcal{H}\}_{\mathrm{PB}}$$

where $\{\mathcal{A}, \mathcal{B}\}_{\mathrm{PB}}$ is the classical ***Poisson Bracket***.

$$\left[p_r, p_s\right] \equiv p_r\, p_s - p_s\, p_r = 0 \qquad \text{[I-15b]}$$

$$\left[q_r, p_s\right] \equiv q_r\, p_s - p_s\, q_r = i\hbar\left\{q_r, p_s\right\}_{\mathrm{PB}} = i\hbar\delta_{rs} \qquad \text{[I-15c]}$$

These relationships provide the foundation for the analogy between quantum and classical mechanics and they show that classical mechanics may be regarded as the limiting case of quantum mechanics when $\hbar$ tends to zero.

THE SCHRÖDINGER OR COORDINATE REPRESENTATION:
The canonical coordinate(s) is (are) diagonal in the Schrödinger representation so that

$$q_1\, q_2\, q_3 \cdots\cdots \left| q_1'\, q_2''\, q_3''' \cdots\cdots \right\rangle = q_1'\, q_2''\, q_3''' \cdots\cdots \left| q_1'\, q_2''\, q_3''' \cdots\cdots \right\rangle \qquad \text{[I-16]}$$

For simplicity, consider a system with a single degree of freedom. For such system any ket $\left|\psi(q)\right\rangle$ has a representative $\psi(q') = \left\langle q' \middle| \psi(q)\right\rangle$ -- *i.e.*

$$\left|\psi(q)\right\rangle = \int d\,q'\, \left|q'\right\rangle\left\langle q'\middle|\psi(q)\right\rangle = \int d\,q'\, \left|q'\right\rangle \psi(q') \qquad \text{[I-17]}$$

The linear operator d/dq acting on this ket generates a new ket which has a representative which is the derivative of the first representative -- *i.e.*

$$\frac{d}{dq}\left|\psi(q)\right\rangle = \left|\frac{d\psi(q)}{dq}\right\rangle \qquad \text{[I-18a]}$$

which means that

$$\left|\frac{d\psi(q)}{dq}\right\rangle = \int dq'\,|q'\rangle\left\langle q'\middle|\frac{d\psi(q)}{dq}\right\rangle = \int dq'\,|q'\rangle\frac{d\psi(q')}{dq'}\,.\text{[9]} \qquad \text{[I-18b]}$$

Now the crucial commutator of q and d/dq may be obtain by the following argument:

$$\begin{aligned}\frac{d}{dq}q|\psi(q)\rangle = \frac{d}{dq}|q\psi(q)\rangle = \left|\frac{dq\psi(q)}{dq}\right\rangle &= \left|q\frac{d\psi(q)}{dq}\right\rangle + |\psi(q)\rangle \\ &= q\frac{d}{dq}|\psi(q)\rangle + |\psi(q)\rangle\end{aligned} \qquad \text{[I-19a]}$$

and, since $|\psi(q)\rangle$ is arbitrary,

$$\frac{d}{dq}q - q\frac{d}{dq} = 1\ . \qquad \text{[I-19b]}$$

Therefore, to be consistent with Equation [I-15c] we take

$$p_r \equiv -i\hbar\frac{\partial}{\partial q_r}\ \ . . \qquad \text{[I-20]}$$

THE MOMENTUM REPRESENTATION:

The canonical momenta are diagonal in the momentum representation[10] so that

$$p_1\,p_2\,p_3\cdots\cdots|p_1'\,p_2''\,p_3'''\cdots\cdots\rangle = p_1'\,p_2''\,p_3'''\cdots\cdots|p_1'\,p_2''\,p_3'''\cdots\cdots\rangle\ . \qquad \text{[I-21]}$$

By using Equations [I-18b] and [I-20], we can show, for one degree of freedom, that the representative of the momentum in the Schrödinger representation is given by

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[9] It is easily shown that $\langle\varphi(q)|\frac{d}{dq} = -\left\langle\frac{d\varphi(q)}{dq}\right|$ which means that $\left\{\frac{d}{dq}\right\}^{\dagger} = -\frac{d}{dq}$ and that $\frac{d}{dq}$ is ***a purely imaginary linear operator***.

[10] The momentum representation is, generally, less useful than the Schrödinger representation. Because of the manifest symmetry between q and p we can now easily show that $q_r = i\hbar\, d/dp_r$.

$$\langle q'|p|p'\rangle = p'\langle q'|p'\rangle = -i\hbar\langle q'|\frac{\partial}{\partial q}|p'\rangle = -i\hbar\frac{\partial}{\partial q'}\langle q'|p'\rangle \qquad [\text{ I-22 }]$$

which has the solution $\langle q'|p'\rangle = c(p')\exp\left(i\,p'\,q'/\hbar\right)$. [I-23]

This connection between the Schrödinger and momentum representations leads to the important insight: the coordinate (or momentum) representative of some arbitrary ket $|A\rangle$ is given by the Fourier components of the momentum (or coordinate) representative -- *i.e.*

$$\langle p'|A\rangle = (2\pi\hbar)^{-1/2}\int_{-\infty}^{+\infty}\exp\left(-i\,p'q'/\hbar\right)\,dq'\,\langle q'|A\rangle \qquad [\text{ I-24a }]$$

$$\langle q'|A\rangle = (2\pi\hbar)^{-1/2}\int_{-\infty}^{+\infty}\exp\left(i\,p'q'/\hbar\right)\,dp'\,\langle p'|A\rangle \qquad [\text{ I-24b }]$$

Application of this transform pair to a *wave packets* in the Schrödinger representation leads directly to the Heisenberg Uncertainty Principle.

A NOTE ON UNITARY TRANSFORMATIONS

Let $\mathcal{U}$ be any linear operator that has a reciprocal $\mathcal{U}^{-1}$. Consider the transformation

$$\alpha^{\mathrm{T}} = \mathcal{U}\,\alpha\,\mathcal{U}^{-1} \qquad [\text{ I-25 }]$$

where α is an arbitrary linear operator. This may be interpreted as expressing a transformation from any linear operator α to a corresponding operator α^{T}. The following properties hold:

a. α^{T} has the same eigenvalues as the corresponding α -- *i.e.* if $\alpha|\alpha'\rangle = \alpha'|\alpha'\rangle$ then

$$\mathcal{U}\,\alpha|\alpha'\rangle = \mathcal{U}\,\alpha\,\mathcal{U}^{-1}\mathcal{U}|\alpha'\rangle = \alpha^{\mathrm{UT}}\,\mathcal{U}|\alpha'\rangle = \alpha'\,\mathcal{U}|\alpha'\rangle\,. \qquad [\text{ I-26a }]$$

b. The fundamental algebraic processes of addition and multiplication are left invariant by the transformation -- *e.g.*,

$$\begin{aligned}(\alpha_1+\alpha_2)^{T} &= \mathcal{U}\,(\alpha_1+\alpha_2)\,\mathcal{U}^{-1} = \mathcal{U}\,\alpha_1\,\mathcal{U}^{-1} + \mathcal{U}\,\alpha_2\,\mathcal{U}^{-1} = \alpha_1^{T} + \alpha_2^{T} \\ (\alpha_1\,\alpha_2)^{T} &= \mathcal{U}\,\alpha_1\,\alpha_2\,\mathcal{U}^{-1} = \mathcal{U}\,\alpha_1\,\mathcal{U}^{-1}\,\mathcal{U}\,\alpha_2\,\mathcal{U}^{-1} = \alpha_1^{T}\,\alpha_2^{T}\end{aligned} \qquad \text{[I-26b]}$$

c. If $\mathcal{U}$ transforms a real α into a real α^{T}

then $\qquad \alpha^{T}\mathcal{U} = \mathcal{U}\,\alpha \qquad \mathcal{U}^{*}\,\alpha^{T} = \alpha\,\mathcal{U}^{*}$

and $\qquad \mathcal{U}^{*}\,\alpha^{T}\,\mathcal{U} = \mathcal{U}^{*}\,\mathcal{U}\,\alpha \qquad \mathcal{U}^{*}\,\alpha^{T}\,\mathcal{U} = \alpha\,\mathcal{U}^{*}\,\mathcal{U}$

therefore $\qquad \mathcal{U}^{*}\,\mathcal{U} = I \qquad \mathcal{U}^{*} = \mathcal{U}^{-1} \qquad$ [I-26c]

Such a transformation is say to be a ***unitary transformation***.

II. REVIEW OF BASIC QUANTUM MECHANICS: DYNAMIC BEHAVIOR OF QUANTUM SYSTEMS:

QUANTUM MECHANICAL EQUATIONS OF MOTION:

To this point our review, we have been concerned with describing the states of a system at one instant of time. The complete dynamical theory must describe, of course, connections between different instants of time.

> "When one makes an observation on the dynamical system, the state of the system gets changed in an unpredictable way, but in between observations causality applies, in quantum mechanics as in classical mechanics, and the system is governed by equations of motion which make the state at one time determined the state at a later time." [10]

Thus, it is only the disturbance caused by the interaction of a system with a measuring device that makes the system's behavior cease to be strictly causal.

SCHRÖDINGER EQUATION OF MOTION:

Consider the time evolution of a particular state of an undisturbed system. To deal with such a dynamical system, we need a linear operator of the form[11]

$$\left|\psi(t)\right\rangle = \mathcal{T}\left(t,t_0\right)\left|\psi(t_0)\right\rangle \qquad \text{[II-1]}$$

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[10] From Section 27 of P. A. M. Dirac, *The Principles of Quantum Mechanics* (Revised fourth edition), Oxford University Press (1967).

[11] This is first member of a class of "displacement " operators that we may treat in a similar fashion. See Section 25 of P. A. M. Dirac, *The Principles of Quantum Mechanics* (Revised fourth edition), Oxford University Press (1967).

Passing to the limit $t \rightarrow t_0$, this operator yields a related linear operator for the time derivative of the state vector with respect to t_0

$$\frac{d}{dt_0}\left|\psi(t_0)\right\rangle = \left\{ \lim_{t \rightarrow t_0} \frac{\mathcal{T}(t,t_0) - 1}{t - t_0} \right\} \left|\psi(t_0)\right\rangle = \mathcal{Op}(t_0)\left|\psi(t_0)\right\rangle \qquad \text{[II-2a]}$$

If it is postulated that $\mathcal{Op}(t) = (i\hbar)^{-1}\,\mathcal{H}(t)$ where $\mathcal{H}(t)$ is the total Hamiltonian (energy) of the system,[12] we obtain the Schrödinger equation of motion in abstract form -- *viz.*

$$i\hbar\frac{d}{dt}\left|\psi(t)\right\rangle = \mathcal{H}(t)\left|\psi(t)\right\rangle \qquad \text{[II-2b]}$$

Equations [II-1] and [II-2b] are consistent if

$$i\hbar\frac{d}{dt}\mathcal{T}(t,t_0) = \mathcal{H}(t)\,\mathcal{T}(t,t_0)\ . \qquad \text{[II-3]}$$

In the **Schrödinger representation**, the abstract **Schrödinger equation of motion** becomes[13]

$$i\hbar\frac{\partial}{\partial t}\psi(\vec{r},t) = \left\langle\vec{r}\right|\mathcal{H}(t)\left|\vec{r}\right\rangle\,\psi(\vec{r},t) \qquad \text{[II-4]}$$

HEISENBERG EQUATION OF MOTION:

In the ***Schrödinger picture***, as outlined above, we picture the states of the undisturbed motion by associating each state with a moving ket, the state at any time corresponding to the ket at that time. In the ***Heisenberg picture*** a unitary transformation is applied which brings to rest the kets corresponding to states of undisturbed motion. In this picture, the appropriate equation of motion is one describing the motion or evolution of linear operators (dynamic variables) -- *viz*.

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[12] There are two justifications of this postulate: (a) analogy with classical mechanics (see Equation [II-6]) and (b) relativistic invariance *vis-a-vis* Equation [I-20].

[13] Since $\left|\psi(t)\right\rangle = \iiint d\vec{r}\,\left|\vec{r}\right\rangle\left\langle\vec{r}\right|\psi(t)\rangle = \iiint d\vec{r}\,\left|\vec{r}\right\rangle\,\psi(\vec{r},t)$

$$\mathcal{O}p_t = \mathcal{T}^{-1}\mathcal{O}p\,\mathcal{T} \quad \text{or} \quad \mathcal{T}\,\mathcal{O}p_t = \mathcal{O}p\,\mathcal{T} \qquad \text{[II-5a]}$$

where $\mathcal{O}p$ is a fixed *Schrödinger dynamic variable* and $\mathcal{O}p_t$ is a time varying *Heisenberg dynamic variable*. Differentiating with respect to time

$$\frac{d\mathcal{T}}{dt}\,\mathcal{O}p_t + \mathcal{T}\,\frac{d\mathcal{O}p_t}{dt} = \mathcal{O}p\,\frac{d\mathcal{T}}{dt} \qquad \text{[II-5b]}$$

and then using Equation [II-3], we obtain the Heisenberg form of the equation of motion as

$$i\hbar\frac{d\mathcal{O}p_t}{dt} = \mathcal{T}^{-1}\mathcal{O}p\,\mathcal{H}\,\mathcal{T} - \mathcal{T}^{-1}\mathcal{H}\,\mathcal{T}\,\mathcal{O}p_t = \left[\mathcal{O}p_t, \mathcal{H}_t\right] \qquad \text{[II-6]}$$

where $\mathcal{H}_t = \mathcal{T}^{-1}\mathcal{H}\,\mathcal{T}$. Note that this equation -- *i.e.* Equation [II-6] -- is consistent with the classical analogy discussed earlier.

ENERGY EIGENVECTOR REPRESENTATION -- THE HEISENBERG REPRESENTATION:

In the Heisenberg picture the stationary states $|E_n\rangle$ correspond to fixed eigenvectors of a time independent Hamiltonian .

CASE 1 -- Time independent Hamiltonian:

To study the time evolution of a given state when the Hamiltonian is time independent, we expand the state vector in terms of these fixed energy eigenvectors and write Equation [II-2b] as

$$i\hbar\frac{d}{dt}\left|\psi(t)\right\rangle = \mathcal{H}\left|\psi(t)\right\rangle = \sum_n \mathcal{H}\left|E_n\right\rangle\left\langle E_n\right|\psi(t)\rangle = \sum_n E_n\left|E_n\right\rangle\left\langle E_n\right|\psi(t)\rangle \qquad [\text{ II-7a }]$$

and
$$i\hbar\frac{\partial}{\partial t}\left\langle E_n\right|\psi(t)\rangle = \sum_{n'} E_{n'}\left\langle E_n\right|E_{n'}\rangle\left\langle E_{n'}\right|\psi(t)\rangle = E_n\left\langle E_n\right|\psi(t)\rangle\ . \qquad [\text{ II-7b }]$$

Therefore

$$\begin{aligned}\left\langle E_n\right|\psi(t)\rangle &= \left\langle E_n\right|\psi(0)\rangle\ \exp\left(\frac{E_n t}{i\hbar}\right) = \left\langle E_n\right|\psi(0)\rangle\ \exp(-i\omega_n t) \\ &= C_n(0)\ \exp(-i\omega_n t)\end{aligned} \qquad [\text{ II-8 }]$$

We may expand an arbitrary wave function in terms of these eigenvectors -- *viz*.

$$\begin{aligned}\psi(\vec{r},t) &= \left\langle \vec{r}\right|\psi(t)\rangle = \sum_n \left\langle \vec{r}\right|E_n\rangle\left\langle E_n\right|\psi(t)\rangle = \sum_n \left\langle E_n\right|\psi(0)\rangle\left\langle \vec{r}\right|E_n\rangle \exp(-i\omega_n t) \\ &= \sum_n C_n(0)\ u_n(\vec{r})\ \exp(-i\omega_n t)\end{aligned} \qquad [\text{ II-9 }]$$

where $u_n(\vec{r}) \equiv \left\langle \vec{r}\right|E_n\rangle$ and the coefficients $C_n(0) = \left\langle E_n\right|\psi(0)\rangle$ are, **in this instance,** of course independent of time.

CASE 2 -- Time dependent Hamiltonian:

Consider now a time dependent Hamiltonian in the form

$$\mathcal{H}(t) = \mathcal{H}_0 + \mathcal{H}_1(t) \qquad [\text{ II-10 }]$$

In this instance, we may write Equation [II-2b] as

$$\begin{aligned}i\hbar\frac{d}{dt}\left|\psi(t)\right\rangle &= \sum_n \mathcal{H}_0\left|E_n\right\rangle\left\langle E_n\right|\psi(t)\rangle + \sum_n \mathcal{H}_1(t)\left|E_n\right\rangle\left\langle E_n\right|\psi(t)\rangle \\ &= \sum_n E_n\left|E_n\right\rangle\left\langle E_n\right|\psi(t)\rangle + \sum_n \mathcal{H}_1(t)\left|E_n\right\rangle\left\langle E_n\right|\psi(t)\rangle\end{aligned} \qquad [\text{ II-11a }]$$

which becomes, on operating from the left with $\langle E_{n'}|$

$$i\hbar\frac{d}{dt}\langle E_{n'}|\psi(t)\rangle = \sum_n E_n\,\langle E_{n'}|E_n\rangle\langle E_n|\,\psi(t)\rangle + \sum_n \langle E_{n'}|\mathcal{H}_1(t)\,|E_n\rangle\langle E_n|\psi(t)\rangle \qquad \text{[II-11b]}$$

If we write $\langle E_n|\psi(t)\rangle = C_n(t)\ \exp(-i\omega_n t)$ -- see Equation [II-8] -- then

$$\left[i\hbar\,\dot{C}_n(t) + E_n\,C_n(t)\right]\exp(-i\omega_n t) = E_n\,C_n(t)\exp(-i\omega_n t) + \sum_{n'}\langle E_n|\mathcal{H}_1(t)|E_{n'}\rangle C_{n'}(t)\exp(-i\,\omega_{n'} t) \qquad \text{[II-11c]}$$

Therefore, when we expand an arbitrary wave function *a la* Equation [II-9], we find

$$\langle\vec{r}|\,\psi(t)\rangle = \sum_n\langle\vec{r}\,|E_n\rangle\langle E_n|\psi(t)\rangle = \sum_n C_n(t)\,\langle\vec{r}\,|E_n\rangle\exp(-i\omega_n t) = \sum_n C_n(t)\ u_n(\vec{r})\ \exp(-i\omega_n t) \qquad \text{[II-12a]}$$

and

$$\dot{C}_n(t) = -\frac{i}{\hbar}\sum_{n'} C_{n'}(t)\,\langle E_n|\mathcal{H}_1(t)\,|E_{n'}\rangle\ \exp(\,i\,\omega_{nn'}t) \qquad \text{[II-12b]}$$

where $\omega_{nn'} = \omega_n - \omega_{n'}$ and

$$\langle E_n|\mathcal{H}_1(t)|E_{n'}\rangle = \iiint d\vec{r}\ u_n(\vec{r})\ \mathcal{H}_1(t)\ \ u_{n'}(\vec{r})\,. \qquad \text{[II-13]}$$

If we write $c_n(t) = C_n(t)\ \exp(-i\,\omega_n t)$, $c_n(t)$ obeys the equation of motion

$$\dot{c}_n(t) = -i\,\omega_n\ c_n(t) - \frac{i}{\hbar}\sum_{n'} c_{n'}(t)\,\langle E_n|\mathcal{H}_1(t)\,|E_{n'}\rangle \qquad \text{[II-14]}$$

TIME DEPENDENT PERTURBATION THEORY:

ELEMENTARY IDEAS -- FIRST ORDER ITERATION:

In set of equations denoted as Equation [II-12b] assume that

$$C_i(0) = 1 \quad \text{and} \quad C_n(0) = 0 \;\; \text{for} \;\; n \neq i \qquad \text{[II-15]}$$

so that in first-order

$$\dot{C}_n(t) \approx \dot{C}_n^{(1)}(t) = -\frac{i}{\hbar}\langle E_n|\mathcal{H}_1(t)|E_i\rangle \exp(i\,\omega_{ni}\,t) \qquad \text{[II-16]}$$

Suppose that

$$\mathcal{H}_1(t) = \mathcal{V}_0 \cos\omega_r t = \mathcal{V}_0 \left\{ \frac{\exp(i\omega_r t) + \exp(-i\omega_r t)}{2} \right\} \qquad \text{[II-17]}$$

for $t \geq 0$, then

$$C_n(t) \approx C_n^{(1)}(t) = -i\frac{\langle E_n|\mathcal{V}_0|E_i\rangle}{2\hbar} \left\{ \frac{\exp[i(\omega_{ni}+\omega_r)t] - 1}{i(\omega_{ni}+\omega_r)} + \frac{\exp[i(\omega_{ni}-\omega_r)t] - 1}{i(\omega_{ni}-\omega_r)} \right\} \qquad \text{[II-18]}$$

CASE 1: $\omega_r = 0$

$$\left|C_n^{(1)}(t)\right|^2 = \frac{\left|\langle E_n|\mathcal{V}_0|E_i\rangle\right|^2}{\hbar^2} \frac{\sin^2(\omega_{ni}\,t/2)}{(\omega_{ni}/2)^2} \qquad \text{[II-19]}$$

which is valid as long as $C_i(t) \approx 1$. Equation [II-18] **shows that transitions are more likely if energy is conserved between initial and final states**.

CASE 2: $\omega_r \neq 0$

In the so called ***rotating-wave approximation*** we neglect the first term in Equation [II-18] so that

$$\left|C_n^{(1)}(t)\right|^2 = \frac{\left|\langle E_n|\mathcal{V}_0|E_i\rangle\right|^2}{4\hbar^2}\frac{\sin^2\left[\left(\omega_{ni}-\omega_r\right)t/2\right]}{\left[\left(\omega_{ni}-\omega_r\right)/2\right]^2} \qquad \text{[II-20]}$$

which is again valid as long as $C_i(t)\approx 1$. This equation **shows that transitions are unlikely unless the resonance condition $\omega_r \approx \omega_{ni}$ is satisfied.**

Let us suppose that we have continuum of energy levels represented by a density of states function $\rho(\omega)$. Therefore the total probability for transition out of the initial state has the approximate value

$$\mathrm{P}_T = \int \rho(\omega)\left|C_n^{(1)}(t)\right|^2 d\omega = \int \rho(\omega)\frac{\left|\mathcal{V}(\omega)\right|^2}{4\hbar^2}\,t^2\,\frac{\sin^2\left[\left(\omega-\omega_r\right)t/2\right]}{\left[\left(\omega-\omega_r\right)t/2\right]^2}\,d\omega \qquad \text{[II-21a]}$$

For $\left(\omega-\omega_r\right)t \ll \pi$

$$\mathrm{P}_T \approx t^2\int\rho(\omega)\frac{\left|\mathcal{V}(\omega)\right|^2}{4\hbar^2}\,d\omega \qquad \text{[II-21b]}$$

so that initial the transition rate -- *i.e.*, $\frac{d\,\mathrm{P}_T}{dt}$ -- is linearly dependent on t. For longer times, it is reasonable to assume that the "frequency-width" of $\rho(\omega)\left|\mathcal{V}(\omega)\right|^2$ is large compared to the inverse of the elapsed time -- or more precisely $\frac{\pi}{t}$ --we may further approximate the total transition probability as

$$\mathrm{P}_T \approx \rho(\omega_r)\frac{\left|\mathcal{V}(\omega_r)\right|^2}{2\hbar^2}\,t\int_{-\infty}^{+\infty}\frac{\sin^2 u}{u^2}\,du \approx \frac{\pi}{2\hbar^2}\rho(\omega_r)\left|\mathcal{V}(\omega_r)\right|^2 t \qquad \text{[II-21c]}$$

which yields the famous **Fermi Golden Rule** for the transition rate

$$\Gamma = \frac{d\,\mathrm{P}_T}{dt} = -\frac{d}{dt}\left|C_i^{(1)}\right|^2 = \frac{\pi}{2\hbar^2}\,\rho(\omega_r)\left|\mathcal{V}(\omega_r)\right|^2 \qquad [\text{ II-22 }]$$

HIGHER-ORDER TIME DEPENDENT PERTURBATION THEORY:

Consider once again perturbation solutions for a Hamiltonian of the form $\mathcal{H}(t) = \mathcal{H}_0 + \mathcal{H}_1(t)$. At outset we transform our equations to the *halfway land* of the **interaction picture** by first rewriting Equation [II-2b] to emphasize that it deals with a state vector in the Schrödinger picture -- *viz*.

$$i\hbar\frac{d}{dt}\left|\psi_S(t)\right\rangle = \mathcal{H}\left|\psi_S(t)\right\rangle \qquad [\text{ II-2b' }]$$

Then we make the unitary transformation

$$\left|\psi_S(t)\right\rangle = \mathcal{T}_0(t)\left|\psi_I(t)\right\rangle = \exp\left(-i\mathcal{H}_0\,t/\hbar\right)\left|\psi_I(t)\right\rangle \qquad [\text{ II-23 }]$$

so that Equation [II-2b'] becomes

$$i\hbar\frac{d\mathcal{T}_0(t)}{dt}\left|\psi_I(t)\right\rangle + i\hbar\,\mathcal{T}_0(t)\frac{d}{dt}\left|\psi_I(t)\right\rangle = \mathcal{T}_0(t)\,\mathcal{H}_0\left|\psi_I(t)\right\rangle + \mathcal{H}_1(t)\,\mathcal{T}_0(t)\left|\psi_I(t)\right\rangle \qquad [\text{ II-24 }]$$

Using a version of Equation [II-3] we obtain the equation of motion for the state in the interaction picture -- *viz*.

$$\frac{d}{dt}\left|\psi_I(t)\right\rangle = -\frac{i}{\hbar}\mathcal{T}_0^{\dagger}(t)\,\mathcal{H}_1(t)\,\mathcal{T}_0(t)\left|\psi_I(t)\right\rangle = -\frac{i}{\hbar}\mathcal{H}_1^{\,I}(t)\left|\psi_I(t)\right\rangle \qquad [\text{ II-25 }]$$

If we write

$$\left|\psi_I(t)\right\rangle = \mathcal{T}_I(t)\left|\psi_I(0)\right\rangle \qquad [\text{ II-26a }]$$

so that

$$\frac{d}{dt}\mathcal{T}_I(t) = -\frac{i}{\hbar}\,\mathcal{H}_1^{\,I}(t)\,\mathcal{T}_I(t) \tag{II-26b}$$

We may formally integrate this equation to obtain

$$\mathcal{T}_I(t) = 1 + \left(-\frac{i}{\hbar}\right)\int_0^t dt'\,\mathcal{H}_1^{\,I}(t')\,\mathcal{T}_I(t') \tag{II-27a}$$

By iterating once we have

$$\mathcal{T}_I(t) = 1 + \left(-\frac{i}{\hbar}\right)\int_0^t dt_1\,\mathcal{H}_1^{\,I}(t_1) + \left(-\frac{i}{\hbar}\right)^2\int_0^t dt_1\,\mathcal{H}_1^{\,I}(t_1)\int_0^{t_1} dt_2\,\mathcal{H}_1^{\,I}(t_2)\,\mathcal{T}_I(t_2) \tag{II-27b}$$

or by successive iterations we have to an arbitrary level of precision

$$\begin{aligned}\mathcal{T}_I(t) = 1 &+ \left(-\frac{i}{\hbar}\right)\int_0^t dt_1\,\mathcal{H}_1^{\,I}(t_1) + \left(-\frac{i}{\hbar}\right)^2\int_0^t dt_1\,\mathcal{H}_1^{\,I}(t_1)\int_0^{t_1} dt_2\,\mathcal{H}_1^{\,I}(t_2)\\ &+ \left(-\frac{i}{\hbar}\right)^3\int_0^t dt_1\,\mathcal{H}_1^{\,I}(t_1)\int_0^{t_1} dt_2\,\mathcal{H}_1^{\,I}(t_2)\int_0^{t_2} dt_3\,\mathcal{H}_1^{\,I}(t_3) + \cdots\cdots\end{aligned} \tag{II-27c}$$

DENSITY OPERATOR (MATRIX):

In classical theory any state of a dynamic system is represented as a point in the *phase space* whose number of dimensions is twice (coordinate and momentum) the number of degrees of freedom in the system. This point will move according to the classical equations of motion. If the state of the system is defined by some probabilistic specification, then we know only the probability that the system may be assigned a given phase point at a particular time. We may envisage the time varying probabilistic specification as a ***fluid*** of density $\rho(q_1, q_2, q_3, \cdots p_1, p_2, p_3, \cdots, t)$ moving through phase space. Each particle of the fluid will move according to the system's equations of motion. The following conservation relationship holds:

$$\frac{\partial \rho}{\partial t} = -\sum_r \left\{ \frac{\partial}{\partial q_r}\left[\rho \frac{dq_r}{dt}\right] + \frac{\partial}{\partial p_r}\left[\rho \frac{dp_r}{dt}\right] \right\} = -\left\{\rho, \mathcal{H}\right\}_{\mathrm{PB}} \qquad [\text{ II-28 }]$$

John von Neumann first introduced a corresponding density function into quantum mechanics. Dirac pointed out that the existence of such a quantum mechanical density is surprising in view of the fact that phase space has no meaning in quantum mechanics since numerical values cannot be assigned simultaneously to the coordinates and momenta. The quantum mechanical density operator (dyadic) is defined as

$$\rho = \sum_m |m\rangle\, P_m \,\langle m| \qquad [\text{ II-29 }]$$

where P_m is the probability of the system being in the state $|m\rangle$.

The equation of motion for the density operator is easily determined from the Schrödinger equation -- *i.e.* Equation [II-2b] -- as

$$\begin{aligned}\frac{\partial \rho}{\partial t} &= \sum_m P_m \left\{ \frac{\partial |\dot{m}\rangle}{\partial t}\langle m| + |m\rangle \frac{\partial \langle \dot{m}|}{\partial t} \right\} \\ &= -\frac{i}{\hbar}\sum_m P_m \left\{ \mathcal{H}\, |m\rangle\langle m| - |m\rangle\langle m| \mathcal{H} \right\} \ . \qquad [\text{ II-30 }] \\ &= -\frac{i}{\hbar}\left[\mathcal{H}, \rho\right]\end{aligned}$$

This equation is, thus, the classical analogy of Equation [II-28]. Let us now express the expectation value of an operator $\mathcal{O}p$ in terms of the density operator -- *viz*.

$$\begin{aligned}\langle \mathcal{O}p \rangle &= \sum_{\psi} P_{\psi} \langle \psi | \mathcal{O}p \; | \psi \rangle = \sum_{\psi} P_{\psi} \sum_{m} \langle \psi | \mathcal{O}p \; | m \rangle \langle m | \psi \rangle \\ &= \sum_{m} \sum_{\psi} P_{\psi} \langle m | \psi \rangle \langle \psi | \mathcal{O}p \; | m \rangle = \sum_{m} \langle m | \rho \, \mathcal{O}p \; | m \rangle \qquad \text{[II-31]} \\ &= \mathrm{tr}\left(\rho \, \mathcal{O}p \right)\end{aligned}$$

Gibbs showed that when a dynamic system is in thermodynamic equilibrium with its surroundings at a given temperature T, the density is give by

$$\rho = (\text{constant}) \; \exp\left[- \mathcal{H} / kT \right] \; . \qquad \text{[II-32a]}$$

This formula is taken over unchanged into quantum mechanics.

$$\rho = (\text{constant}) \; \exp\left[- \mathcal{H} / kT \right] \qquad \text{[II-32b]}$$

APPENDIX - TIME INDEPENDENT PERTURBATION THEORY

Suppose that we have a Hamiltonian $\mathcal{H} = \mathcal{B} + \mathcal{S}$ where $\mathcal{B}$ is a big part and $\mathcal{S}$ is a small part. Of course, we want the solution to the complete eigenvalue problem - *i.e.*

$$\mathcal{H}\left|\mathcal{H}^n\right\rangle = \mathcal{H}^n\left|\mathcal{H}^n\right\rangle \qquad \text{[A-1]}$$

To obtain an approximate solution we expand the eigenvalues and eigenvectors in a series of terms of ***increasing smallness*** - *viz.*

$$\mathcal{H}^n = \mathcal{H}_0^n + \mathcal{H}_1^n + \mathcal{H}_2^n + \mathcal{H}_3^n + \cdots \qquad \text{[A-2a]}$$

$$\left|\mathcal{H}^n\right\rangle = \left|\mathcal{H}_0^n\right\rangle + \left|\mathcal{H}_1^n\right\rangle + \left|\mathcal{H}_2^n\right\rangle + \cdots \qquad \text{[A-2b]}$$

$$\left\langle\mathcal{H}^n\right| = \left\langle\mathcal{H}_0^n\right| + \left\langle\mathcal{H}_1^n\right| + \left\langle\mathcal{H}_2^n\right| + \cdots \qquad \text{[A-2c]}$$

Let us first consider normalization - *i.e.*

$$\left\langle\mathcal{H}^n\middle|\mathcal{H}^n\right\rangle = \left(\left\langle\mathcal{H}_0^n\right| + \left\langle\mathcal{H}_1^n\right| + \left\langle\mathcal{H}_2^n\right| + \cdots\right)\left(\left|\mathcal{H}_0^n\right\rangle + \left|\mathcal{H}_1^n\right\rangle + \left|\mathcal{H}_2^n\right\rangle + \cdots\right) = 1 \qquad \text{[A-3a]}$$

Compare terms of ***equal smallness*** so that

$$\left\langle\mathcal{H}^n\middle|\mathcal{H}^n\right\rangle = \left\langle\mathcal{H}_0^n\middle|\mathcal{H}_0^n\right\rangle = 1 \qquad \text{[A-3b]}$$

$$\left\langle\mathcal{H}_0^n\middle|\mathcal{H}_1^n\right\rangle + \left\langle\mathcal{H}_1^n\middle|\mathcal{H}_0^n\right\rangle = 0 \qquad \text{[A-3c]}$$

$$\left\langle\mathcal{H}_0^n\middle|\mathcal{H}_2^n\right\rangle + \left\langle\mathcal{H}_1^n\middle|\mathcal{H}_1^n\right\rangle + \left\langle\mathcal{H}_2^n\middle|\mathcal{H}_0^n\right\rangle = 0 \qquad etc. \qquad \text{[A-3d]}$$

From Equation [A-3c] we see that we may take $\left\langle\mathcal{H}_1^n\middle|\mathcal{H}_0^n\right\rangle = 0$ without introducing error. Next we write an expanded version of the eigenvalue equation [A-1]

$$\begin{aligned}&(\mathcal{B}+\mathcal{S})\left(\left|\mathcal{H}_0^n\right\rangle+\left|\mathcal{H}_1^n\right\rangle+\left|\mathcal{H}_2^n\right\rangle+\cdots\right)\\&\quad=\left(\mathcal{H}_0^n+\mathcal{H}_1^n+\mathcal{H}_2^n+\mathcal{H}_3^n+\cdots\right)\left(\left|\mathcal{H}_0^n\right\rangle+\left|\mathcal{H}_1^n\right\rangle+\left|\mathcal{H}_2^n\right\rangle+\cdots\right)\end{aligned}\qquad\text{[A-4a]}$$

Again compare terms of ***equal smallness*** so that

$$\mathcal{B}\left|\mathcal{H}_0^n\right\rangle=\mathcal{H}_0^n\left|\mathcal{H}_0^n\right\rangle\qquad\text{[A-4b]}$$

$$\mathcal{S}\left|\mathcal{H}_0^n\right\rangle+\mathcal{B}\left|\mathcal{H}_1^n\right\rangle=\mathcal{H}_0^n\left|\mathcal{H}_1^n\right\rangle+\mathcal{H}_1^n\left|\mathcal{H}_0^n\right\rangle\qquad\text{[A-4c]}$$

$$\mathcal{B}\left|\mathcal{H}_2^n\right\rangle+\mathcal{S}\left|\mathcal{H}_1^n\right\rangle=\mathcal{H}_0^n\left|\mathcal{H}_2^n\right\rangle+\mathcal{H}_1^n\left|\mathcal{H}_1^n\right\rangle+\mathcal{H}_2^n\left|\mathcal{H}_0^n\right\rangle\quad etc.\qquad\text{[A-4d]}$$

Operate through Equation [A-4c] with $\left\langle\mathcal{H}_0^n\right|$ and using Equations [A-3b] and [A-4b] we find

$$\mathcal{H}_1^n=\left\langle\mathcal{H}_0^n\right|\mathcal{S}\left|\mathcal{H}_0^n\right\rangle\qquad\text{[A-5]}$$

Operate through Equation [A-4c] with $\left\langle\mathcal{H}_0^m\right|$ and using again Equations [A-3b] and [A-4b] we find

$$\left\langle\mathcal{H}_0^m\right|\mathcal{S}\left|\mathcal{H}_0^n\right\rangle=\left(\mathcal{H}_0^m-\mathcal{H}_0^n\right)\left\langle\mathcal{H}_0^m\middle|\mathcal{H}_1^n\right\rangle\qquad\text{[A-6a]}$$

or we have the **representative**

$$\left\langle\mathcal{H}_0^m\middle|\mathcal{H}_1^n\right\rangle=\frac{\left\langle\mathcal{H}_0^m\right|\mathcal{S}\left|\mathcal{H}_0^n\right\rangle}{\left(\mathcal{H}_0^m-\mathcal{H}_0^n\right)}\qquad\text{[A-6b]}$$

and

$$\left|\mathcal{H}_1^n\right\rangle=\sum_m\left|\mathcal{H}_0^m\right\rangle\left\langle\mathcal{H}_0^m\middle|\mathcal{H}_1^n\right\rangle=\sum_{m\neq n}\left|\mathcal{H}_0^m\right\rangle\frac{\left\langle\mathcal{H}_0^m\right|\mathcal{S}\left|\mathcal{H}_0^n\right\rangle}{\left(\mathcal{H}_0^m-\mathcal{H}_0^n\right)}\qquad\text{[A-6c]}$$

Operate through Equation [A-4d] with $\left\langle\mathcal{H}_0^n\right|$ and, of course, using Equations [A-3b] and [A-4b] we find

$$\left\langle \mathcal{H}_0^n \middle| \mathcal{S} \middle| \mathcal{H}_1^n \right\rangle = \mathcal{H}_1^n \left\langle \mathcal{H}_0^n \middle| \mathcal{H}_1^n \right\rangle + \mathcal{H}_2^n = \mathcal{H}_2^n \qquad [\text{ A-7a }]$$

or

$$\begin{aligned} \mathcal{H}_2^n &= \left\langle \mathcal{H}_0^n \middle| \mathcal{S} \middle| \mathcal{H}_1^n \right\rangle = \sum_{m \neq n} \left\langle \mathcal{H}_0^n \middle| \mathcal{S} \middle| \mathcal{H}_0^m \right\rangle \frac{\left\langle \mathcal{H}_0^m \middle| \mathcal{S} \middle| \mathcal{H}_0^n \right\rangle}{\left(\mathcal{H}_0^m - \mathcal{H}_0^n \right)} \\ &= \sum_{m \neq n} \frac{\left| \left\langle \mathcal{H}_0^m \middle| \mathcal{S} \middle| \mathcal{H}_0^n \right\rangle \right|^2}{\left(\mathcal{H}_0^m - \mathcal{H}_0^n \right)} \end{aligned} \qquad [\text{ A-7b }]$$

III. REVIEW OF BASIC QUANTUM MECHANICS: TWO-LEVEL QUANTUM SYSTEMS:

The literature of quantum optics and laser spectroscopy abounds with discussions of the two-level (two-state) system. This emphasis comes about because the interaction of such systems with the electromagnetic field may be treated in great detail to obtain valuable analytic results and, hopefully, the analysis of two-level systems generates insights that may be extended to more realistic situations. Fortunately, there are several important instances in which the application of the two-level model provides a very good approximation to a more complete theory. In the following, we label the upper level of the system by the letter **a** and the lower by the letter **b**. From Equation [I-12a] we write, specifically, the wave function of the two level system as

$$\psi(\vec{r}\,,t) = \langle \vec{r}\,|\,\psi(t)\rangle = C_a(t)\, u_a(\vec{r})\, \exp(-i\,\omega_a t) + C_b(t)\, u_b(\vec{r})\, \exp(-i\,\omega_b t) \qquad \text{[III-1]}$$

where we know from Equation [I-12b] that the time varying coefficients satisfy, in general, the following equations:

$$\dot{C}_a(t) = -\frac{i}{\hbar}\Big\{C_a(t)\,\langle E_a|\mathcal{H}_1(t)\,|E_a\rangle + C_b(t)\,\langle E_a|\mathcal{H}_1(t)\,|E_b\rangle\, \exp(\;i\,\omega_{ab} t)\Big\} \qquad \text{[III-2a]}$$

$$\dot{C}_b(t) = -\frac{i}{\hbar}\Big\{C_b(t)\,\langle E_b|\mathcal{H}_1(t)\,|E_b\rangle + C_a(t)\,\langle E_b|\mathcal{H}_1(t)\,|E_a\rangle\, \exp(\;i\,\omega_{ba} t)\Big\} \qquad \text{[III-2b]}$$

If we take the interaction to be the electric dipole interaction with an applied electric field we may write

$$\mathcal{H}_1(t) = -\vec{\mathbf{p}}_S\;\; \vec{\mathbf{E}}\left(\vec{\mathbf{R}}_S\,, t\right) = -e\,\vec{\mathbf{r}}\;\; \vec{\mathbf{E}}\left(\vec{\mathbf{R}}_S\,, t\right) \qquad \text{[III-3a]}$$

where $\vec{\mathbf{R}}_S$ denotes the position of the center of the two-level system or atom.[14] Thus we write

$$\langle E_i | \mathcal{H}_1(t) | E_j \rangle = \mathcal{V}_{ij} = -\langle E_i |\, e\,\vec{\mathbf{r}}\, | E_j \rangle \; \vec{\mathbf{E}}\left(\vec{\mathbf{R}}_S, t\right) \qquad \text{[III-3b]}$$

In all but the most bizarre circumstances we may use persuasive symmetry arguments to reason that

$$\langle E_i |\, e\,\vec{\mathbf{r}}\, | E_i \rangle \equiv 0$$

Thus Equations [III-2] reduce to

$$\dot{C}_a(t) = -\frac{i}{\hbar} C_b(t)\, \mathcal{V}_{ab} \; \exp\left(i\omega_{ab} t\right) \qquad \text{[III-4a]}$$

$$\dot{C}_b(t) = -\frac{i}{\hbar} C_a(t)\, \mathcal{V}_{ab} \; \exp\left(-i\omega_{ab} t\right) \qquad \text{[III-4b]}$$

where $\mathcal{V}_{ab} = -\langle E_a |\, e\,\vec{\mathbf{r}}\, | E_b \rangle \; \vec{\mathbf{E}}\left(\vec{\mathbf{R}}_S, t\right) = - \quad \mathrm{E}\left(\vec{\mathbf{R}}_S, t\right)$.

RABI FLOPPING -- WITHOUT DAMPING:

For an oscillatory applied field

$$\mathcal{V}_{ab} = -| \quad \mathrm{E}_o | \cos \omega_r\, t = -\tfrac{1}{2} \quad \mathrm{E}_o \exp\left(-i\omega_r\, t\right) + c.c. \qquad \text{[III-5]}$$

we see, in the ***rotating-wave approximation***, that

$$\begin{aligned} \dot{C}_a(t) &= \frac{i}{2\hbar} \quad \mathrm{E}_o\, C_b(t) \, \exp\left[\, i \left(\omega_{ab} - \omega_r\right) t\right] \\ &= \frac{i}{2}\, \Omega_{\mathbf{o}}^{\mathbf{R}} \; C_b(t) \exp\left[\, i \left(\omega_{ab} - \omega_r\right) t\right] \end{aligned} \qquad \text{[III-6a]}$$

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[14] The use of this form of interaction needs considerable elaboration, but we defer that discussion until later.

$$\begin{aligned} \dot{C}_b(t) &= \frac{i}{2\hbar} \quad \mathrm{E_o} \; C_a(t) \exp\left[-i\left(\omega_{ab}-\omega_r\right)t\right] \\ &= \frac{i}{2}\Omega_{\mathbf{o}}^{\mathbf{R}} \; C_a(t) \; \exp\left[-i\left(\omega_{ab}-\omega_r\right)t\right] \end{aligned} \qquad \text{[III-6b]}$$

where $\Omega_{\mathbf{o}}^{\mathbf{R}} \equiv \quad \mathrm{E_o}/\hbar$ defines the so called **Rabi flopping frequency** which is, of course, a measure of the strength of the electromagnetic interaction**.** Clearly, the coupling terms have maximum effect when the frequency of the applied field is resonant with the level splitting. In most treatments the ***frequency detuning*** of the field is expressed as $\delta\omega = \omega_{ab} - \omega_r$ and the system's wave function - *i.e.* Equation [III-1] - is written in terms of slightly modified time varying coefficients by transforming to the ***rotating frame of reference*** - *viz.*

$$C_a(t) = \tilde{C}_a(t) \exp\left(i \; \tfrac{1}{2}\delta\omega \, t\right) \qquad \text{[III-7a]}$$

$$C_b(t) = \tilde{C}_b(t) \exp\left(-i \; \tfrac{1}{2}\delta\omega \, t\right) \qquad \text{[III-7b]}$$

$$\psi(\vec{r}, t) = \tilde{C}_a(t) \; \exp\left[i\left(\tfrac{1}{2}\delta\omega - \omega_a\right) t\right] u_a(\vec{r}) + \tilde{C}_b(t) \; \exp\left[i\left(-\tfrac{1}{2}\delta\omega - \omega_b\right) t\right] u_b(\vec{r}) \qquad \text{[III-7c]}$$

The coupling terms in this rotating frame of reference become

$$\dot{\tilde{C}}_a(t) = \tfrac{1}{2}\, i \left\{-\delta\omega \tilde{C}_a(t) + \Omega_{\mathbf{o}}^{\mathbf{R}} \; \tilde{C}_b(t)\right\} \qquad \text{[III-8a]}$$

$$\dot{\tilde{C}}_b(t) = \tfrac{1}{2}\, i \left\{ \delta\omega \tilde{C}_b(t) + \Omega_{\mathbf{o}}^{\mathbf{R}} \; \tilde{C}_a(t)\right\} \qquad \text{[III-8b]}$$

or in matrix form

$$\frac{d}{dt}\,\tilde{\mathbf{C}}(t) = \frac{d}{dt}\begin{pmatrix} \tilde{C}_a(t) \\ \tilde{C}_b(t) \end{pmatrix} = \frac{i}{2}\begin{pmatrix} \delta\omega & \Omega_{\mathbf{o}}^{\mathbf{R}} \\ \Omega_{\mathbf{o}}^{\mathbf{R}} & \delta\omega \end{pmatrix}\begin{pmatrix} \tilde{C}_a(t) \\ \tilde{C}_b(t) \end{pmatrix} = \frac{i}{2}\,\mathbf{M} \;\; \tilde{\mathbf{C}}(t) \; . \qquad \text{[III-8c]}$$

We look for a solution in the form $\tilde{\mathbf{C}}(t) = \tilde{\mathbf{C}}(0)\,\exp\left(\tfrac{1}{2}\,i\,\Omega^{\mathbf{R}}\,t\right)$ where $\Omega^{\mathbf{R}}$ is the generalization of the Rabi flopping frequency. Therefore, the condition

$$\det\left[\mathbf{M} - \Omega^{\mathbf{R}}\,\mathbf{I}\right] = 0$$

yields the ***generalized Rabi flopping frequency***

$$\Omega^{\mathbf{R}} = \sqrt{\delta\omega^2 + \left|\Omega^{\mathbf{R}}_{\mathbf{o}}\right|^2} \qquad \text{[III-9a]}$$

and the general **non-damped** time evolving wave function

$$\begin{pmatrix}\tilde{C}_a(t)\\ \tilde{C}_b(t)\end{pmatrix} = \begin{pmatrix} \cos\left(\tfrac{1}{2}\Omega^{\mathbf{R}}\,t\right) - \dfrac{i\,\delta\omega}{\Omega^{\mathbf{R}}}\sin\left(\tfrac{1}{2}\Omega^{\mathbf{R}}\,t\right) & \dfrac{i\,\Omega^{\mathbf{R}}_{\mathbf{o}}}{\Omega^{\mathbf{R}}}\sin\left(\tfrac{1}{2}\Omega^{\mathbf{R}}\,t\right) \\ \dfrac{i\,\Omega^{\mathbf{R}}_{\mathbf{o}}}{\Omega^{\mathbf{R}}}\sin\left(\tfrac{1}{2}\Omega^{\mathbf{R}}\,t\right) & \cos\left(\tfrac{1}{2}\Omega^{\mathbf{R}}\,t\right) + \dfrac{i\,\delta\omega}{\Omega^{\mathbf{R}}}\sin\left(\tfrac{1}{2}\Omega^{\mathbf{R}}\,t\right) \end{pmatrix} \begin{pmatrix}\tilde{C}_a(0)\\ \tilde{C}_b(0)\end{pmatrix} \qquad \text{[III-9b]}$$

RABI FLOPPING -- WITH DAMPING:

Neglected interactions (e.g. spontaneous emission, collisions, thermal fluctuations) limit lifetime of a state of a two-level system. One class of lifetime limiting interactions may be described **phenomenologically** by adding decay terms to the equations of motion - *i.e.* to Equations [III-8] - as follows:

$$\dot{\tilde{C}}_a(t) = -\tfrac{1}{2}\left(\gamma_a + i\,\delta\omega\right)\tilde{C}_a(t) + \tfrac{1}{2}\,i\,\Omega^{\mathbf{R}}_{\mathbf{o}}\,\tilde{C}_b(t) \qquad \text{[III-10a]}$$

$$\dot{\tilde{C}}_b(t) = -\tfrac{1}{2}\left(\gamma_b - i\,\delta\omega\right)\tilde{C}_b(t) + \tfrac{1}{2}\,i\,\Omega^{\mathbf{R}}_{\mathbf{o}}\,\tilde{C}_a(t) \qquad \text{[III-10b]}$$

or

$$\frac{d}{dt}\,\tilde{\mathbf{C}}(\mathbf{t}) = \frac{d}{dt}\begin{pmatrix}\tilde{C}_a(t)\\ \tilde{C}_b(t)\end{pmatrix} = \frac{i}{2}\begin{pmatrix}\left(\delta\omega - i\gamma_a\right) & \Omega^{\mathbf{R}}_{\mathbf{o}} \\ \Omega^{\mathbf{R}}_{\mathbf{o}} & \left(\delta\omega + i\gamma_b\right)\end{pmatrix}\begin{pmatrix}\tilde{C}_a(t)\\ \tilde{C}_b(t)\end{pmatrix} = \frac{i}{2}\,\mathbf{M}\;\;\tilde{\mathbf{C}}(t) \qquad \text{[III-10c]}$$

Again we look for a solution in the form $\tilde{\mathbf{C}}(t) = \tilde{\mathbf{C}}(0)\,\exp\left(\tfrac{1}{2}\,i\,\Lambda t\right)$ so that

$$\det\left[\mathbf{M} - \Lambda\,\mathbf{I}\right] = 0$$

or

$$\Lambda^2 + i\left(\gamma_b + \gamma_a\right)\Lambda - \left[\left(\delta\omega - i\gamma_a\right)\left(\delta\omega + i\gamma_b\right) + \left|\Omega_{\mathbf{o}}^{\mathbf{R}}\right|^2\right] = 0\,.$$

Therefore

$$\Lambda = i\,\tfrac{1}{2}\left(\gamma_a + \gamma_b\right) \pm \sqrt{\left[\delta\omega - \tfrac{1}{2}\,i\left(\gamma_a - \gamma_b\right)\right]^2 + \left|\Omega_{\mathbf{o}}^{\mathbf{R}}\right|^2} \qquad \text{[III-11a]}$$

-- where the we refer to $\gamma_{ab} = \tfrac{1}{2}\left(\gamma_a + \gamma_b\right)$ as the **average decay rate constant** and $\overline{\Omega}^{\mathbf{R}} = \sqrt{\left[\delta\omega - \tfrac{1}{2}\,i\left(\gamma_a - \gamma_b\right)\right]^2 + \left|\Omega_{\mathbf{o}}^{\mathbf{R}}\right|^2}$ as the **generalized complex Rabi flopping frequency** -- and the general time evolving wave function -- in the rotation frame -- may be written

$$\begin{pmatrix} \tilde{C}_a(t) \\ \tilde{C}_b(t) \end{pmatrix} = \begin{pmatrix} \cos\left(\tfrac{1}{2}\,\overline{\Omega}^{\mathbf{R}}\,t\right) - \dfrac{\left[\tfrac{1}{2}\left(\gamma_a - \gamma_b\right) + i\,\delta\omega\right]}{\overline{\Omega}^{\mathbf{R}}}\sin\left(\tfrac{1}{2}\,\overline{\Omega}^{\mathbf{R}}\,t\right) & \dfrac{i\,\Omega_{\mathbf{o}}^{\mathbf{R}}}{\overline{\Omega}^{\mathbf{R}}}\sin\left(\tfrac{1}{2}\,\overline{\Omega}^{\mathbf{R}}\,t\right) \\ \dfrac{i\,\Omega_{\mathbf{o}}^{\mathbf{R}}}{\overline{\Omega}^{\mathbf{R}}}\sin\left(\tfrac{1}{2}\,\overline{\Omega}^{\mathbf{R}}\,t\right) & \cos\left(\tfrac{1}{2}\,\overline{\Omega}^{\mathbf{R}}\,t\right) + \dfrac{\tfrac{1}{2}\left(\gamma_a - \gamma_b\right) + i\,\delta\omega}{\overline{\Omega}^{\mathbf{R}}}\sin\left(\tfrac{1}{2}\,\overline{\Omega}^{\mathbf{R}}\,t\right) \end{pmatrix} \exp\left(-\tfrac{1}{2}\,\gamma_{ab}\,t\right) \begin{pmatrix} \tilde{C}_a(0) \\ \tilde{C}_b(0) \end{pmatrix} \qquad \text{[III-11b]}$$

DENSITY MATRIX TREATMENT OF A TWO-LEVEL SYSTEMS:

Recall from Equation [II-30]

$$\dot{\rho} = \frac{i}{\hbar}\left[\rho, \mathcal{H}\right] = \frac{i}{\hbar}\left\{\rho\,\mathcal{H} - \mathcal{H}\,\rho\right\}$$

Using $|a\rangle\langle a| + |b\rangle\langle b| = 1$ we may write

$$\dot{\rho} = \frac{i}{\hbar}\left\{\rho\left(|a\rangle\langle a| + |b\rangle\langle b|\right)\mathcal{H} - \mathcal{H}\left(|a\rangle\langle a| + |b\rangle\langle b|\right)\rho\right\} \qquad \text{[III-12a]}$$

If $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1$, then

$$\begin{aligned}\dot{\rho} = \frac{i}{\hbar}\Big\{\rho\,\big[E_a\,|a\rangle\langle a| + E_b\,|b\rangle\langle b| + |a\rangle\langle a|\,\mathcal{H}_1 + |b\rangle\langle b|\,\mathcal{H}_1\big] \\ - \big[E_a\,|a\rangle\langle a| + E_b\,|b\rangle\langle b| + \mathcal{H}_1\,|a\rangle\langle a| + \mathcal{H}_1\,|b\rangle\langle b|\big]\,\rho\Big\}\end{aligned} \qquad \text{[III-12b]}$$

Writing matrix elements -- *i.e.* representatives -- of the density operator

$$\langle a|\,\dot{\rho}\,|a\rangle = \frac{i}{\hbar}\Big\{\langle a|\,\rho\,|b\rangle\langle b|\,\mathcal{H}_1\,|a\rangle - \langle a|\,\mathcal{H}_1\,|b\rangle\langle b|\,\rho\,|a\rangle\Big\} \qquad \text{[III-13a]}$$

$$\langle b|\,\dot{\rho}\,|b\rangle = \frac{i}{\hbar}\Big\{\langle b|\,\rho\,|a\rangle\langle a|\,\mathcal{H}_1\,|b\rangle - \langle b|\,\mathcal{H}_1\,|a\rangle\langle a|\,\rho\,|b\rangle\Big\} \qquad \text{[III-13b]}$$

$$\begin{aligned}\langle a|\,\dot{\rho}\,|b\rangle = \frac{i}{\hbar}\Big\{-\big(E_a - E_b\big)\,\langle a|\,\rho\,|b\rangle + \big[\langle a|\,\rho\,|a\rangle - \langle b|\,\rho\,|b\rangle\big]\,\langle a|\,\mathcal{H}_1\,|b\rangle \\ + \langle a|\,\rho\,|b\rangle\big[\langle b|\,\mathcal{H}_1\,|b\rangle - \langle a|\,\mathcal{H}_1\,|a\rangle\big]\Big\}\end{aligned} \qquad \text{[III-13c]}$$

We may again assume, by symmetry arguments, that $\langle a|\,\mathcal{H}_1\,|a\rangle = \langle b|\,\mathcal{H}_1\,|b\rangle = 0$ so that the equations of motion for the elements of the density reduce to

$$\dot{\rho}_{bb} - \dot{\rho}_{aa} = \big[i\,\hbar^{-1}\,2\,\mathcal{V}_{ab}\,\rho_{ba} + c.c.\big] \qquad \text{[III-14a]}$$

$$\dot{\rho}_{ab} = -\,i\,\omega_{ab}\,\rho_{ab} + i\,\hbar^{-1}\,\mathcal{V}_{ab}\big[\rho_{aa} - \rho_{bb}\big] \qquad \text{[III-14b]}$$

To introduce an element of reality, we add to these equations a pair of the most ***intuitively satisfying*** damping terms (a useful attribute of density matrix formulations) -- *viz.*

$$\dot{\rho}_{bb} - \dot{\rho}_{\rho aa} = \big[i\,\hbar^{-1}\,2\mathcal{V}_{ab}\,\rho_{ba} + c.c.\big] - \Gamma\,\Big[\big(\rho_{bb} - \rho_{aa}\big) - \big(\rho_{bb} - \rho_{aa}\big)_0\Big] \qquad \text{[III-15a]}$$

$$\dot{\rho}_{ab} = -\,i\,\omega_{ab}\,\rho_{ab} + i\,\hbar^{-1}\mathcal{V}_{ab}\left[\rho_{aa} - \rho_{bb}\right] - \gamma\,\rho_{ab} \qquad \text{[III-15b]}$$

Transform these equations to **a rotating frame** by taking $\rho_{ab} = \tilde{\rho}_{ab}\exp\left(-\,i\,\omega_r\ t\right)$ where $\tilde{\rho}_{ab}$ is assumed to be a slowly varying function of time which satisfies the equations of motion

$$\dot{\tilde{\rho}}_{ab} = -\,i\,\delta\omega\tilde{\rho}_{ab} + i\,\hbar^{-1}\mathcal{V}_{ab}\exp\left(\,i\,\omega_r\ t\right)\left[\rho_{aa} - \rho_{bb}\right] - \gamma\,\tilde{\rho}_{ab} \qquad \text{[III-16a]}$$

$$\begin{aligned}\rho_{bb} - \rho_{aa} = {} & \left[i\,\hbar^{-1}\,2\mathcal{V}_{ab}\ \exp\left(\,i\omega_r\ t\right)\tilde{\rho}_{ba} + c.c.\right] \\ & - \Gamma\left[\left(\rho_{bb} - \rho_{aa}\right) - \left(\rho_{bb} - \rho_{aa}\right)_0\right]\end{aligned} \qquad \text{[III-16b]}$$

With $\mathcal{V}_{ab} = -\left[\tfrac{1}{2}\hbar\,\Omega_{\mathbf{o}}^{\mathbf{R}}\ \exp\left(-i\omega_r t\right) + c.c.\right]$ and ignoring terms proportional to $\exp\left(\pm\,i\ 2\omega_r\ t\right)$ -- *i.e.* **the rotating wave approximation** - we find that

$$\dot{\tilde{\rho}}_{ab} = -\,i\left(\delta\omega - i\gamma\right)\tilde{\rho}_{ab} - i\left(\tfrac{1}{2}\Omega_{\mathbf{o}}^{\mathbf{R}}\right)\ \left[\rho_{aa} - \rho_{bb}\right] \qquad \text{[III-17a]}$$

$$\dot{\rho}_{bb} - \dot{\rho}_{aa} = \left[-i\,\Omega_{\mathbf{o}}^{\mathbf{R}}\,\tilde{\rho}_{ba} + c.c.\right] - \Gamma\left[\left(\rho_{bb} - \rho_{aa}\right) - \left(\rho_{bb} - \rho_{aa}\right)_0\right] \qquad \text{[III-17b]}$$

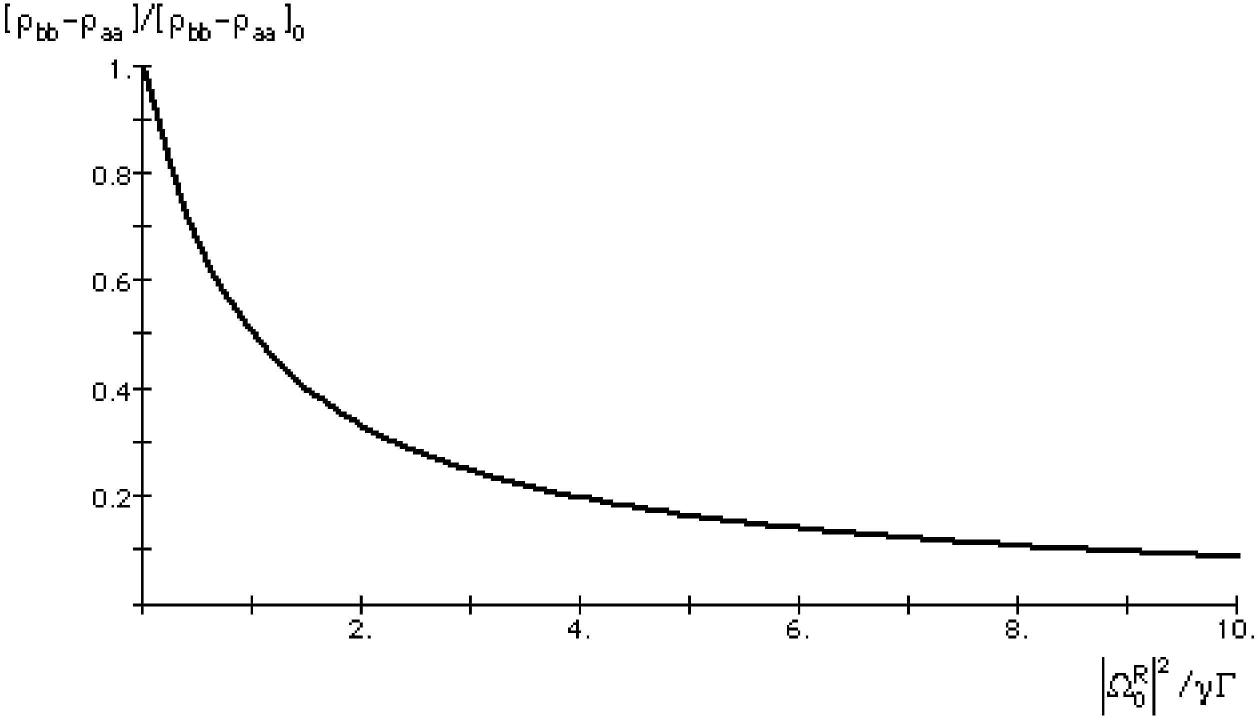
Steady state behavior**:** If we take all time derivatives in these equations equal to zero, we obtain

$$\tilde{\rho}_{ab} = \frac{i\,\Omega_{\mathbf{o}}^{\mathbf{R}}\left[\rho_{bb} - \rho_{aa}\right]}{2\left(\,i\,\delta\omega + \gamma\right)} = \frac{\Omega_{\mathbf{o}}^{\mathbf{R}}\ \left(\,\delta\omega + i\,\gamma\right)\left[\rho_{bb} - \rho_{aa}\right]}{2\left(\delta\omega^2 + \gamma^2\right)} \qquad \text{[III-18a]}$$

$$\left(\rho_{bb} - \rho_{aa}\right) = \left(\rho_{bb} - \rho_{aa}\right)_0\left[\frac{\delta\omega^2 + \gamma^2}{\delta\omega^2 + \gamma^2\left(1 + \left|\Omega_{\mathbf{o}}^{\mathbf{R}}\right|^2/\Gamma\gamma\right)}\right] \qquad \text{[III-18b]}$$

or

$$\tilde{\rho}_{ab} = \frac{\tfrac{1}{2}\,\Omega_o^R \left[\,\delta\omega + i\,\gamma\right]\left(\rho_{bb} - \rho_{aa}\right)_0}{\delta\omega^2 + \gamma^2\left(1 + \left|\Omega_o^R\right|^2 \big/ \Gamma\gamma\right)} \qquad [\text{ III-18c }]$$



We have graphed Equation [III-18b] at resonance - *i.e.* $\delta\omega = 0$ - and, from the following expression, we see how the oscillatory polarization is **saturated** at high electromagnetic powers. In general, the **macroscopic polarization** is then given by[15]

---

[15] That is to say

$$\langle \mathbf{P} \rangle = \left(\mathbf{P}\,\rho\right)_{aa} + \left(\mathbf{P}\,\rho\right)_{bb} = \mathbf{P}_{aa}\,\rho_{aa} + \mathbf{P}_{ab}\,\rho_{ba} + \mathbf{P}_{ba}\,\rho_{ab} + \mathbf{P}_{bb}\,\rho_{bb} = \mathbf{P}_{ab}\,\rho_{ba} + \mathbf{P}_{ba}\,\rho_{ab}$$

since $\mathbf{P}_{aa} = \mathbf{P}_{bb} = 0$ by symmetry.

If we graph Equation [III-18b] at resonance - *i.e.* $\delta\omega = 0$ - we see how the oscillatory polarization is **saturated** at high electromagnetic powers - *viz*

$$P = N\,\langle\text{dipole moment of system}\rangle = N\left[\quad \rho_{ba} + \text{c.c.}\right] \qquad \text{[III-19a]}$$

$$P = \frac{N \quad \Omega_{o}^{R}\ \left(\delta\omega - i\,\gamma\right)\left[\rho_{bb} - \rho_{aa}\right]}{2\left(\delta\omega^2 + \gamma^2\right)}\ \exp\left(i\,\omega_r\ t\right) + \text{c.c.} \qquad \text{[III-19b]}$$

$$P = \frac{N|\quad|^2\left[\ \delta\omega - i\,\gamma\right]\left[\rho_{bb} - \rho_{aa}\right]_0}{2\hbar\left[\delta\omega^2 + \gamma^2\left(1 + \left|\Omega_{o}^{R}\right|^2/\Gamma\gamma\right)\right]}\ \mathrm{E_o}\exp\left(i\,\omega_r\ t\right) + \text{c.c} \qquad \text{[III-19c]}$$

THE VECTOR MODEL OF THE DENSITY MATRIX:

There is a set of **arguments by analogy** which is exceedingly valuable in treating transient excitation problems in optics. The basis for the analogy lies in the fact that Equations [III-16] are identical in form to the **famous Bloch equations of magnetic resonance**.[16] If we make a transformation to a rotating frame -- *i.e.*, $\mathrm{M}_{\pm} = \tilde{\mathrm{M}}_{\pm}\ \exp\left(\mp i\,\omega_r\ t\right)$ -- the Bloch equations take on the form

---

[16] Recall from the theory of magnetic resonance

$$d\vec{\mathbf{M}}/dt = \gamma_{\text{mag}}\ \vec{\mathbf{M}} \times \vec{\mathbf{H}}$$

where γ_{mag} is the **gyromagnetic ratio**. This equation of motion is greatly simplified if it is written in terms of the circular polarizations $\mathrm{M}_{\pm} = \mathrm{M}_x \pm i\,\mathrm{M}_y$ and $\mathrm{H}_{\pm} = \mathrm{H}_x \pm i\,\mathrm{H}_y$ -- *viz.*

$$\dot{\mathrm{M}}_{\pm} = \mp i\ \gamma_{\text{mag}}\ \mathrm{M}_{\pm}\ \mathrm{H}_z \pm i\,\gamma_{\text{mag}}\ \mathrm{M}_z\ \mathrm{H}_{\pm}$$

and

$$\mathrm{M}_z = i\,\frac{\gamma_{\text{mag}}}{2}\left[\mathrm{M}_+\mathrm{H}_- - \mathrm{M}_-\mathrm{H}_+\right]$$

To these Bloch added the phenomenological longitudinal (thermal) relaxation time T_1 and transverse

$$\dot{\tilde{\mathrm{M}}}_\pm = \mp i\,\delta\omega\,\tilde{\mathrm{M}}_\pm \pm i\,\gamma_{\mathrm{mag}}\,\tilde{\mathrm{M}}_z\,\exp\left(\pm i\,\omega_r\,t\right)\mathrm{H}_\pm - \tilde{\mathrm{M}}_\pm\big/T_2 \qquad [\text{ III-20a }]$$

$$\dot{\mathrm{M}}_z = \left[i\,\frac{\gamma_{\mathrm{mag}}}{2}\,\mathrm{H}_-\,\exp\left(-i\,\omega_r\,t\right)\mathrm{M}_+ + c.c.\right] - \left(\mathrm{M}_z - \mathrm{M}_{z0}\right)\big/T_1 \qquad [\text{ III-20b }]$$

where $\delta\omega = \omega_r - \gamma_{\mathrm{mag}}\,\mathrm{H}_z$. By comparing these Bloch equations with Equations [III-16] and [III-17] we see that we have identical problems if we make the identifications

$$\tilde{\mathrm{M}}_+ \Leftrightarrow \tilde{\rho}_{ab} \qquad [\text{ III-21a }]$$

$$\tilde{\mathrm{M}}_z \Leftrightarrow \rho_{bb} - \rho_{aa} \qquad [\text{ III-21b }]$$

$$\gamma_{\mathrm{mag}}\,\mathrm{H}_z \Leftrightarrow \omega_{ab} \qquad [\text{ III-21c }]$$

$$\gamma_{\mathrm{mag}}\,\mathrm{H}_+ \Leftrightarrow \hbar^{-1}\,\mathcal{V}_{ab} = -\tfrac{1}{2}\,\Omega_{\mathbf{o}}^{\mathbf{R}} \qquad [\text{ III-21d }]$$

$$1/T_1 = \Gamma \qquad [\text{ III-21e }]$$

$$1/T_2 = \gamma \qquad [\text{ III-21f }]$$

In other words we have the equivalent equation of motion

$$\dot{\vec{\mathbf{M}}} = \vec{\mathbf{M}} \times \vec{\Omega}_{\mathrm{eff}} - \gamma\left(\vec{\mathbf{M}} - \hat{\mathbf{z}}\,M_z\right) - \Gamma\,\hat{\mathbf{z}}\,M_z \qquad [\text{ III-22 }]$$

where the ***effective Rabi precession field*** $\vec{\Omega}_{\mathrm{eff}}$ is given by

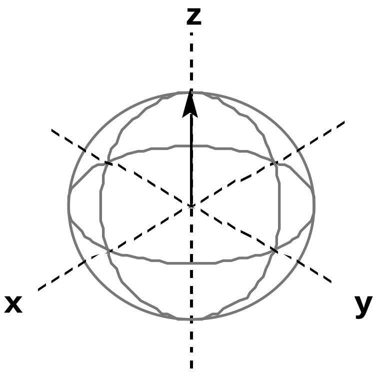
$$\vec{\Omega}_{\mathrm{eff}} = \Omega_{\mathbf{o}}^{\mathbf{R}}\,\hat{\mathbf{x}} - \delta\omega\,\hat{\mathbf{z}} \qquad [\text{ III-23 }]$$

---

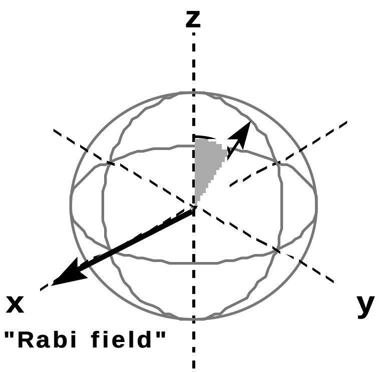
dephasing time T_2 so that

$$\dot{\mathrm{M}}_\pm = \mp i\,\gamma_{\mathrm{mag}}\,\mathrm{M}_\pm\,\mathrm{H}_z \pm i\,\gamma_{\mathrm{mag}}\,\mathrm{M}_z\,\mathrm{H}_\pm - \mathrm{M}_\pm\big/T_2$$

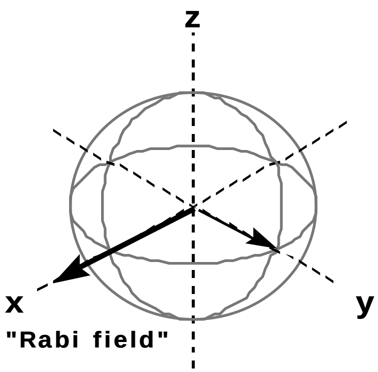
$$\dot{\mathrm{M}}_z = i\,\frac{\gamma_{\mathrm{mag}}}{2}\left[\mathrm{M}_+\mathrm{H}_- - \mathrm{M}_-\mathrm{H}_+\right] - \left(\mathrm{M}_z - \mathrm{M}_{z0}\right)\big/T_1$$



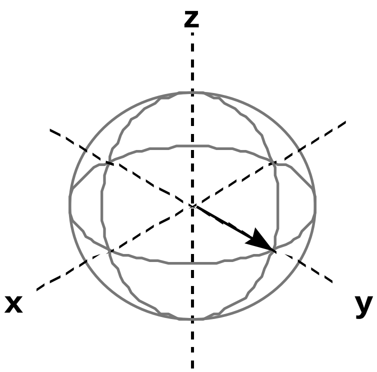
Initial state of system



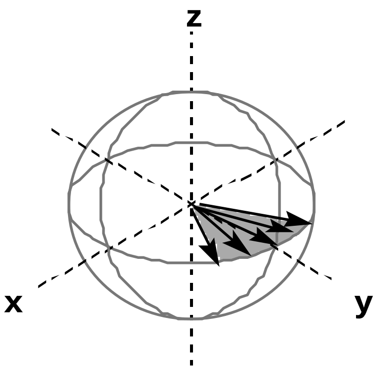
Effect of transient excitation
- precession of state vector



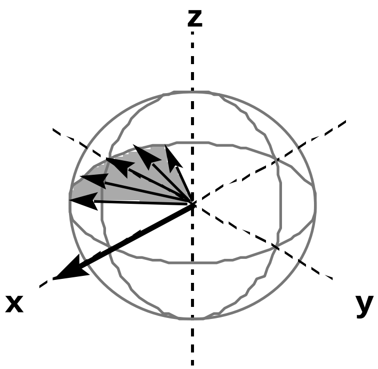
State after 90° pulse



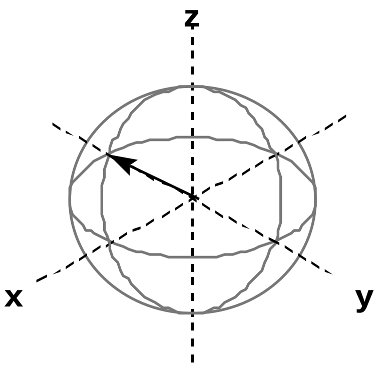
"Rabi field" off
-start of "free decay" period



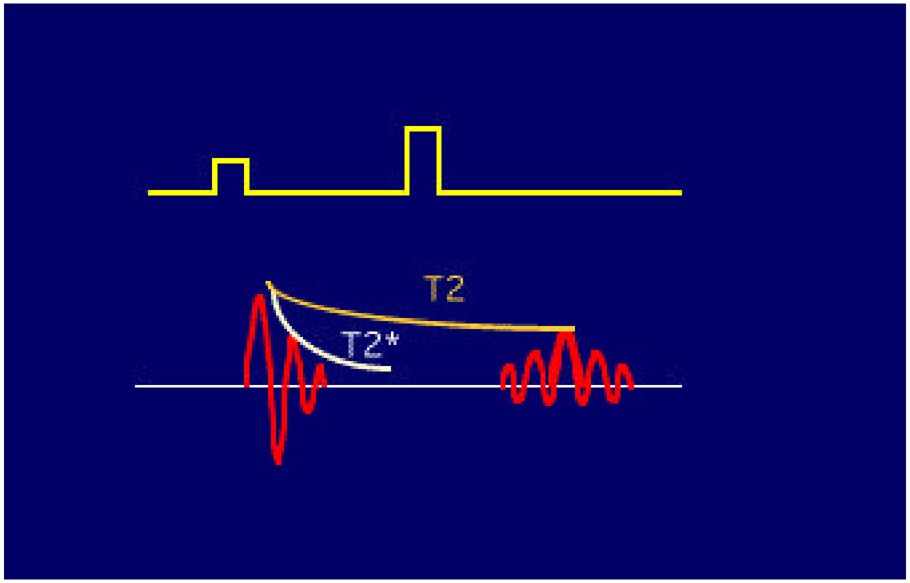
Effect ot inhomogeneities
in level splitting



State of system after 180° pulse



"Echo" state


T2
T2*

IV. REVIEW OF BASIC QUANTUM MECHANICS: THE THERMAL RADIATION FIELD

PLANCK'S RADIATION LAW FOR THERMAL SOURCES:[16]

To set the stage for subsequent discussions of laser physics and quantization of the electromagnetic field we briefly explore the earliest, seminal notions in the quantum theory of light. As we all remember, in 1900 Planck found that he could account for the measured spectral distribution of radiation from a thermal source by postulating that the energies of a ***certain set*** of harmonic oscillators are **quantized!** Drawing on Planck's success, Einstein in 1905 was able to show that the extraordinary features of photoelectric effect could be explained by hypothesizing the ***corpuscularity*** of the electromagnetic field. The crowning triumph of early quantum optics is Einstein's amazingly simple, phenomenological theory of 1917 which provided a quantitative basis for analyzing the absorption and emission of light by atoms.

In treating thermal sources the basic assumption is that emitted radiation is a sample of the total electromagnetic field -- *viz.* all of the modes of a resonator -- in thermal equilibrium with its material environment -- *viz.* the walls of the resonator. In the traditional treatment of the theory of ***black-body radiation***, a particular thermodynamic system is assumed -- *viz.*, a hollow resonator which is a cube of length **L** with has perfectly conducting walls.[17] Thus, to satisfy boundary conditions at the walls, the electric field associated with a particular mode of the cavity is given by

---

[16] This section and some parts of the following section draw heavily upon discussions in Rodney Loudon's *The Quantum Theory of Light* (2nd edition), Oxford (1983).

[17] The detail results of black-body radiation theory are indeed sensitive to the assumed character of the thermodynamic systemas will be demonstration presently.

$$
\begin{aligned}
E_x^{\nu_x,\,\nu_y,\,\nu_z}\left(\vec{\mathbf{r}},\mathbf{t}\right) &= E_x^0\left(k_{\nu_x,\,\nu_y,\,\nu_z}\right)\cos\left(\pi\nu_x\; x/L\right)\sin\left(\pi\nu_y\; y/L\right)\sin\left(\pi\nu_z\; z/L\right)\exp\left(i\,\omega_{\nu_x\nu_y\nu_z}\; t\right)\\
E_y^{\nu_x,\,\nu_y,\,\nu_z}\left(\vec{\mathbf{r}},\mathbf{t}\right) &= E_y^0\left(k_{\nu_x,\,\nu_y,\,\nu_z}\right)\sin\left(\pi\nu_x\; x/L\right)\cos\left(\pi\nu_y\; y/L\right)\sin\left(\pi\nu_z\; z/L\right)\exp\left(i\,\omega_{\nu_x\nu_y\nu_z}\; t\right)\quad [\text{ IV-1 }]\\
E_z^{\nu_x,\,\nu_y,\,\nu_z}\left(\vec{\mathbf{r}},\mathbf{t}\right) &= E_z^0\left(k_{\nu_x,\,\nu_y,\,\nu_z}\right)\sin\left(\pi\nu_x\; x/L\right)\sin\left(\pi\nu_y\; y/L\right)\cos\left(\pi\nu_z\; z/L\right)\exp\left(i\,\omega_{\nu_x\nu_y\nu_z}\; t\right)
\end{aligned}
$$

where $\left\{\nu_x,\nu_y,\nu_z\right\}$ is a set of **positive** integers. To satisfy the homogeneous Helmhotz equation we must have

$$
k^2_{\nu_x\nu_y\nu_z} = \left(\pi\nu_x/L\right)^2 + \left(\pi\nu_y/L\right)^2 + \left(\pi\nu_z/L\right)^2 = \left(\omega_{\nu_x\nu_y\nu_z}/c\right)^2 \qquad [\text{ IV-2 }]
$$

The ***cycle-averaged*** value of the stored energy **density** associated with the particular mode is given by

$$
\begin{aligned}
W\left(k_{\nu_x\nu_y\nu_z}\right) &= \frac{1}{4V}\int_{\text{cavity}}\left[\varepsilon_0\left|\vec{\mathbf{E}}^{\nu_x,\,\nu_y,\,\nu_z}\left(\vec{\mathbf{r}},\mathbf{t}\right)\right|^2 + \mu_0\left|\vec{\mathbf{H}}^{\nu_x,\,\nu_y,\,\nu_z}\left(\vec{\mathbf{r}},\mathbf{t}\right)\right|^2\right]dV\\
&= \frac{1}{2V}\int_{\text{cavity}}\left[\varepsilon_0\left|\vec{\mathbf{E}}^{\nu_x,\,\nu_y,\,\nu_z}\left(\vec{\mathbf{r}},\mathbf{t}\right)\right|^2\right]dV = \frac{1}{16}\,\varepsilon_0\left|\vec{\mathbf{E}}^0\left(k_{\nu_x\nu_y\nu_z}\right)\right|^2
\end{aligned}
\qquad [\text{ IV-3 }]
$$

For a thermal source, the most significant experimentally measurable object is the noise spectrum -- *i.e.,* the frequency distribution of the stored energy density. To obtain this distribution, we take the energy density in the frequency range between ω and $\omega + d\omega$ -- *viz.*

$$
\begin{aligned}
W(\omega)\; d\omega &= \sum_{\{\nu_x,\,\nu_y,\,\nu_z\}} W\left(k_{\nu_x\nu_y\nu_z}\right)\qquad \left\{\text{with } k_{\nu_x\nu_y\nu_z} \text{ between } \omega/c \text{ and } \left(\omega+d\omega\right)/c\right\}\\
&= \left\langle W\left(k_{\nu_x\nu_y\nu_z}\right)\right\rangle \times \left[\begin{matrix}\text{Number of modes with frequencies}\\ \text{between } \omega \text{ and } \omega + d\omega\end{matrix}\right]
\end{aligned}
\qquad [\text{ IV-4 }]
$$

Thus the ***density of states*** is defined as

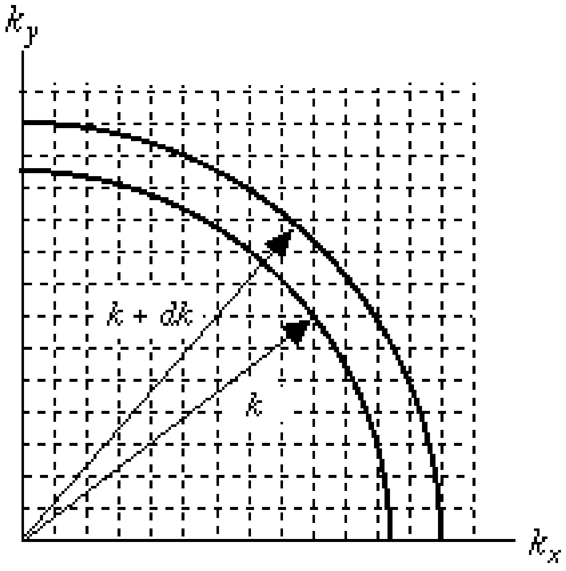
$$\rho_k\, dk \equiv \begin{bmatrix} \text{Number of modes with frequencies} \\ \text{between } \omega \text{ and } \omega + d\omega \end{bmatrix} \qquad \text{[IV-5a]}$$

In the frequency range where $\lambda << 2L$, the following ***k***-space argument holds:

$$\rho_k\, dk \approx \begin{bmatrix}\text{Two polarization}\\ \text{states per } \vec{\mathbf{k}} \text{ state}\end{bmatrix}\begin{bmatrix}\text{Fraction of shell}\\ \text{with vaid states}\end{bmatrix}\begin{bmatrix}\dfrac{\text{Volume in shell}}{\text{Volume of state}} = \text{Number of } \vec{\mathbf{k}} \text{ states within shell}\end{bmatrix}$$

$$\approx 2\times\left(\frac{1}{8}\right)\left[\frac{4\pi\, k^2\, dk}{(\pi/L)^3}\right] = V\left[\frac{\omega^2\, d\omega}{c^3\, \pi^2}\right] \equiv \rho_\omega d\omega \qquad \text{[IV-5b]}$$

This extremely important density-of-states construction may be visualized most elegantly in two dimension.[18]



Cut through shell in **k**-space.

---

[18] For use in later discussions, we note here that by this same argument the 2D density-of-states is found to be $\rho_\omega(2D) = A\omega/\pi c^2$ and the 1D value $\rho_\omega(1D) = L/\pi c$.

Following the traditional (Rayleigh-Jeans) argument, we identify a resonator mode as a harmonic oscillator[19] and take its average energy to be the classical thermodynamic value for a system with two degrees of freedom -- *i.e.* $\left\langle W\left(k_{\nu_x\nu_x\nu_x}\right)\right\rangle_{\text{thermal}} = k_{\text{B}}T/V$ Thus, we obtain the famous *Rayleigh-Jeans radiation law* (which impels us to worry about an ***ultra-violet catastrophe***.)

$$W_{\text{T}}(\omega)\,d\omega = kT\left[\frac{\omega^2\,d\omega}{c^3\,\pi^2}\right] \qquad \text{[IV-6]}$$

Planck's quantization hypothesis:

Following Planck we set

$$W\left(k_{\nu_x\nu_y\nu_z}\right) = \frac{1}{16}\,\varepsilon_0\left|\mathrm{E}^0\left(k_{\nu_x\nu_y\nu_z}\right)\right|^2 = \mathcal{E}_n/V = \left[\hbar\omega\left(n+\tfrac{1}{2}\right)\right]/V \qquad \text{[IV-7]}$$

where n = 0, 1, 2,…… Assuming that the resonator mode is in thermal equilibrium with its environment, we may use the Boltzmann probability factor

$$\exp\left(-\beta\,\mathcal{E}_n\right) = \exp\left(-\mathcal{E}_n/k_{\text{B}}T\right)$$

to find the probability P_n that the mode (read oscillator) is thermally excited to an energy $\mathcal{E}_n$

$$P_n = \frac{\exp\left(-\beta\,\mathcal{E}_n\right)}{\sum_n \exp\left(-\beta\,\mathcal{E}_n\right)} = \frac{\exp\left(-\beta\,n\hbar\,\omega\right)}{\sum_n \exp\left(-\beta\,n\hbar\,\omega\right)} = \exp\left(-\beta\,n\,\hbar\,\omega\right)\left\{1-\exp\left(-\beta\,\hbar\,\omega\right)\right\} \qquad \text{[IV-8]}$$

The thermal mean value of n is obtained in a similar manner [20] -- *viz.*

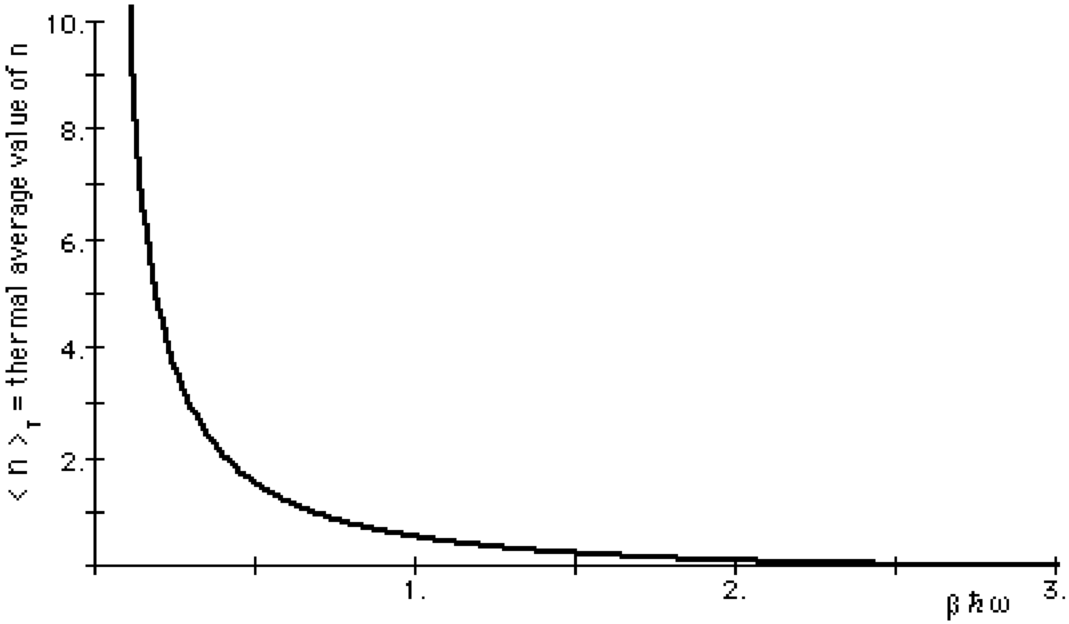
---

[19] We justify this assertion in detail later: it suffices to note here, in anticipation of that later discussion, that a harmonic oscillator and an electromagnetic mode have analogous Hamiltonians.

[20] It is also easy to show that thermal mean-square fluctuation in n is given by $\langle\Delta n\rangle_{\text{T}} = \sqrt{\langle n\rangle_{\text{T}}^2 + \langle n\rangle_{\text{T}}}$.

$$
\begin{aligned}
\langle n \rangle_{\mathrm{T}} &= \sum_n n\, P_n = \frac{\sum_n n \exp(-\beta\, n\, \hbar\, \omega)}{\sum_n \exp(-\beta\, n\, \hbar\, \omega)} \\
&= -\{\partial/\partial(\beta\, \hbar\, \omega)\} \ln\left[\sum_n \exp(-\beta\, n\, \hbar\, \omega)\right] \\
&= -\{\partial/\partial(\beta\, \hbar\, \omega)\} \ln\left[\{1 - \exp(-\beta\, \hbar\, \omega)\}^{-1}\right] \\
&= \frac{1}{\exp(\beta\, \hbar\, \omega) - 1}
\end{aligned}
\qquad [\text{ IV-9 }]
$$

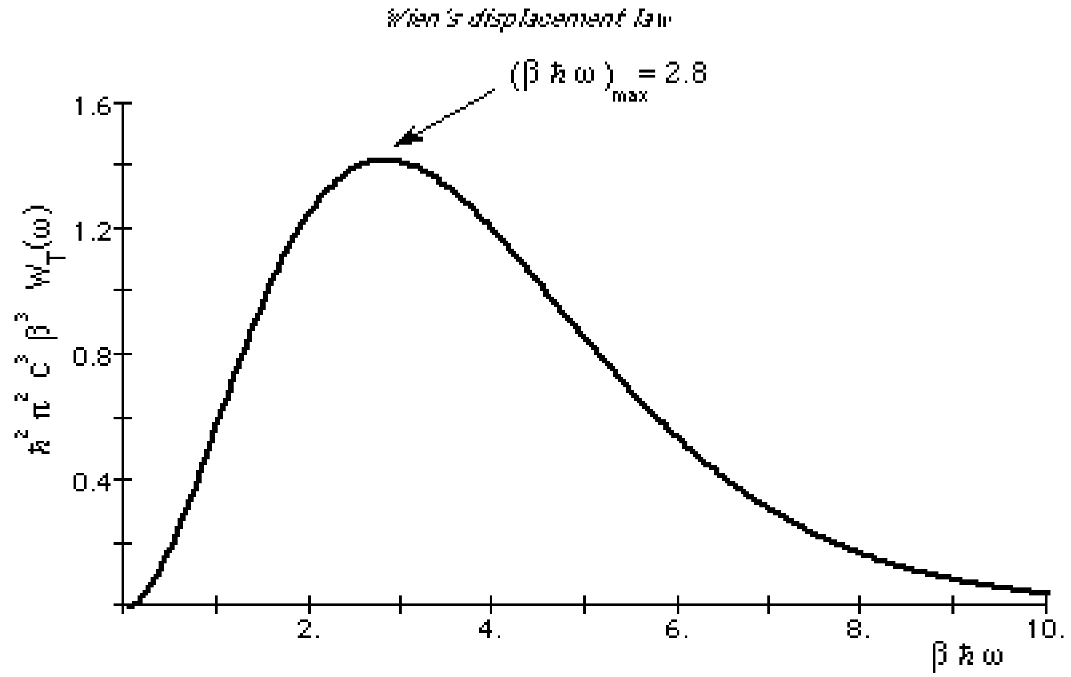
which may be plotted



Equation [IV-9] taken in conjunction with Equations [IV-5b] and [IV-7] leads then directly to the *Planck radiation law*

$$\begin{aligned} W_{\mathrm{T}}(\omega)\,d\omega &= \left\langle n+\tfrac{1}{2}\right\rangle_{\text{thermal}} \left[\frac{\hbar\,\omega^{3}\;d\omega}{c^{3}\,\pi^{2}}\right] \\ &= \left[\frac{1}{\exp(\beta\,\hbar\,\omega)-1}+\frac{1}{2}\right]\left[\frac{(\beta\,\hbar\,\omega)^{3}}{\hbar^{2}\,\pi^{2}\,c^{3}\,\beta^{3}}\right]d\omega \end{aligned} \qquad \text{[IV-10]}$$

From this expression we see that the Rayleigh-Jeans radiation law is correct in the limit $k_{\mathrm{B}}T >> \hbar\,\omega$, but, to our great relief, Planck has staved off the ultraviolet catastrophe. We may then plot this famous and an exceedingly important result. (From the particular point view of laser physics, the Planck radiation law gives a measure of the background radiative **thermal noise spectrum.**)



EINSTEIN'S PHENOMENOLOGICAL THEORY OF RADIATIVE PROCESSES -- Einstein's "A" and "B" coefficients:

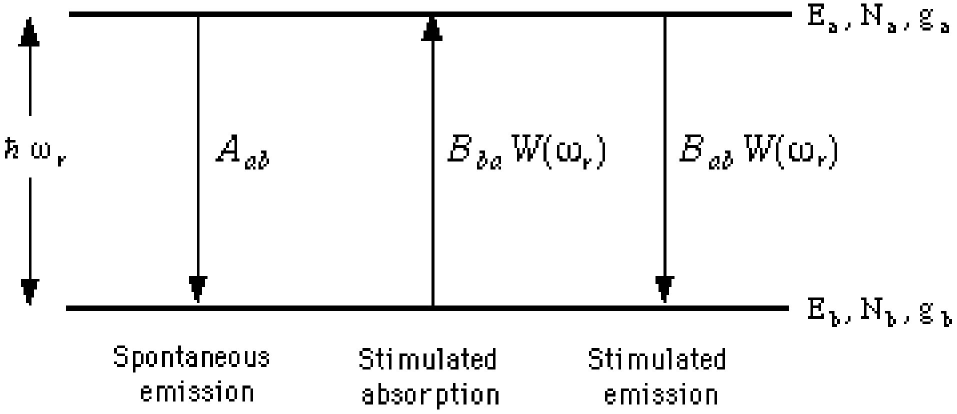
We return to the two-level system and, with Einstein, make some physically reasonable (early quantum mechanical) postulates concern the absorption and emission of light. Consider the two-level system shown below. Einstein's postulates[21] are embodied in a set of rate equations for the level populations- *viz*.

---

[21] Using our semiclassical theory of matter-radiation interaction, we can easily show that $B_{ba} = \frac{\pi|\quad|^2}{3\,\varepsilon_0\,\hbar^2}$,

but, of course, Einstein did not have that well-developed theory at hand. While the stimulated emission and absorption processes are reasonably intuitive, it is the introduction of the spontaneous emission process which flows from a profound understanding of the processes by which thermal equilibrium is achieved.

$$dN_b/dt = N_a A_{ab} - N_b B_{ba} W(\omega_r) + N_a B_{ab} W(\omega_r)$$
$$dN_a/dt = -N_a A_{ab} + N_b B_{ba} W(\omega_r) - N_a B_{ab} W(\omega_r) \qquad [\text{ IV-11 }]$$



It particularly useful to examine the results of these equations under conditions -- *viz*.

General equilibrium or steady-state condition:

$$N_a A_{ab} - N_b B_{ba} W(\omega_r) + N_a B_{ab} W(\omega_r) = 0 \qquad [\text{ IV-12a }]$$

Thermal equilibrium condition:

$$W_T(\omega) = A_{ab}\left[(N_b/N_a) B_{ba} - B_{ab}\right]^{-1} \qquad [\text{ IV-12b }]$$

Of course, in the latter case we use fact that the level populations N_a and N_b at thermal equilibrium are related by Boltzmann's law -- *i.e.*

$$N_b/N_a = \left[g_b \exp(-\beta E_b)\right] / \left[g_a \exp(-\beta E_a)\right] = (g_b/g_a) \exp(\beta \hbar\omega_{ab}) \qquad [\text{ IV-13 }]$$

so that

$$W_T(\omega) = A_{ab}\left[(g_b/g_a) \exp(\beta \hbar\omega) B_{ba} - B_{ab}\right]^{-1} \qquad [\text{ IV-14 }]$$

Thus, by comparison with Equation [IV-10], we see that

$$\left(g_b / g_a\right) B_{ba} = B_{ab} \qquad \text{[IV-15a]}$$

and

$$\left[\frac{\hbar\omega^3}{\pi^2 c^3}\right] B_{ab} = A_{ab} \qquad \text{[IV-15b]}$$

and, by comparison with Equation [IV-9], we see that

$$B_{ab}\, W_{\mathrm{T}}(\omega) = A_{ab} \left\langle n \right\rangle_{\mathrm{T}} \qquad \text{[IV-16a]}$$

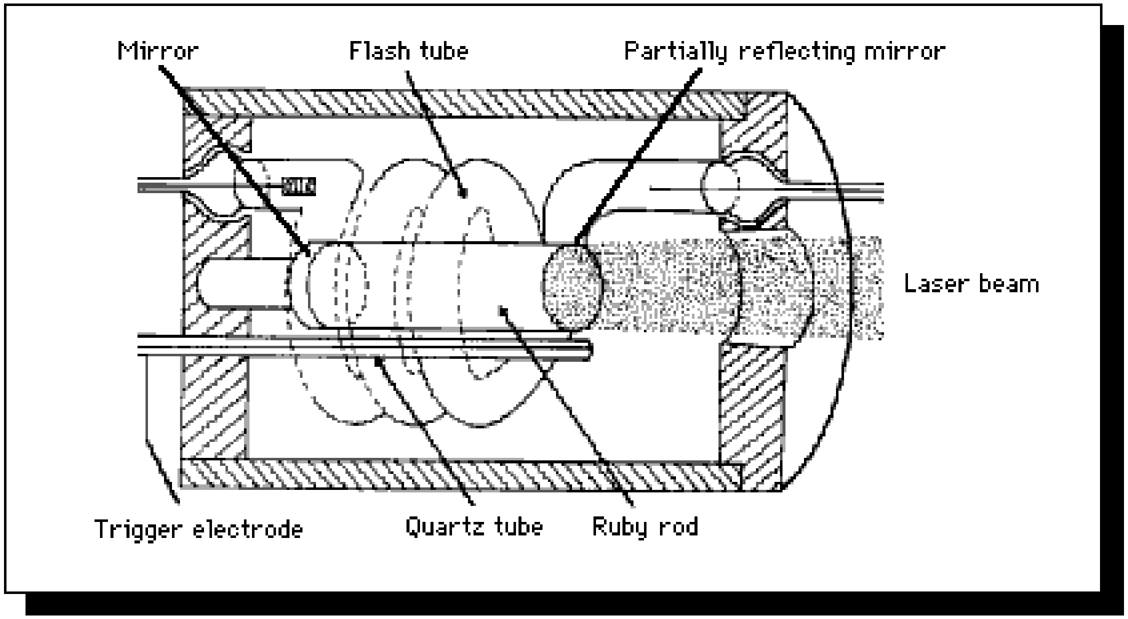
or

$$\frac{\text{The rate of spontaneous emission}}{\text{The rate of stimulated emission}} = \frac{A_{ab}}{B_{ab} W_{\mathrm{T}}(\omega)} = \frac{1}{\left\langle n \right\rangle_{\mathrm{T}}} = \exp(\beta\hbar\omega) - 1 \qquad \text{[IV-16b]}$$

V. ELEMENTARY LASER MODELS -- RATE EQUATION APPROACH:

POPULATION INVERSION AND "LASERING" IN A THREE-LEVEL SYSTEM

Obviously, the trick in building masers and lasers has been to find means to "invert" the equilibrium population of states.[22] First conceived by Charles Townes in 1951, Gordon, Zeiger and Townes in 1954 demonstrated the first ammonia beam maser. Degenerate quantum states of the ammonia molecule (NH_3) are split by the tunneling of the nitrogen atom across the plane formed by the three hydrogen atoms. In the Columbia maser, thermal populations of molecules in the split ground state (23,800 MHz) are spatially separated by an electrostatic field to achieve population inversion in the maser resonant cavity. While this two-level system was clearly seminal, it was the multi-level paradigm of



THE CONFIGURATION OF THEODORE MAIMAN'S RUBY LASER (1960)

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[22] Recently the subject of lasing has become of considerable interest. See, for Thomas Mossberg's very interesting Web Seminar on the subject (http://decryptor.uoregon.edu/~mosswww/moss-seminar.html).

Bloembergen's three-level ruby maser which galvanized the search for possible laser systems.

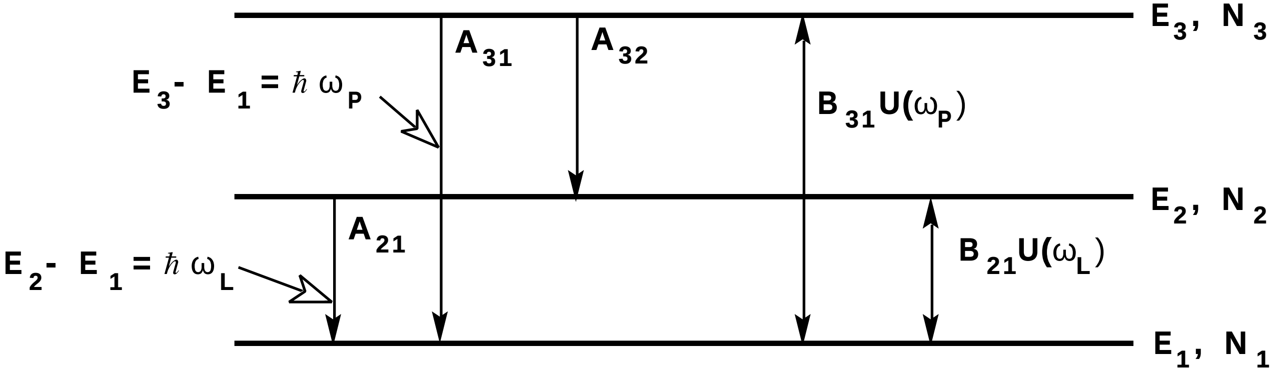
To quote from *A Laser Tutorial* (http://members.aol.com/WSRNet/tut/t1.htm)

> "Finding substances in which a population inversion can be set up is central to the development of new kinds of lasers. The first material used was synthetic ruby. Ruby is crystalline alumina (Al_2O_3) in which a small fraction of the Al^{3+} ions have been replaced by chromium ions, Cr^{3+}. It is the chromium ions that give rise to the characteristic pink or red colour of ruby and it is in these ions that a population inversion is set up in a ruby laser. In a ruby laser, a rod of ruby is irradiated with the intense flash of light from xenon-filled flashtubes. Light in the green and blue regions of the spectrum is absorbed by chromium ions, raising the energy of electrons of the ions from the ground state level to one of the broad F bands of levels. Electrons in the F bands rapidly undergo non-radiative transitions to the two metastable E levels. A non-radiative transition does not result in the emission of light; the energy released in the transition is dissipated as heat in the ruby crystal. The metastable levels are unusual in that they have a relatively long lifetime of about 4 milliseconds (4×10^{-3} s), the major decay process being a transition from the lower level to the ground state. This long lifetime allows a high proportion (more than a half) of the chromium ions to build up in the metastable levels so that a population inversion is set up between these levels and the ground state level. This population inversion is the condition required for stimulated emission to overcome absorption and so give rise to the amplification of light. In an assembly of chromium ions in which a population inversion has been set up, some will decay spontaneously to the ground state level emitting red light of wavelength 694.3 nm in the process. This light can then interact with other chromium ions that are in the metastable levels causing them to emit light of the same wavelength by stimulated emission. As each stimulating photon leads to the emission of two photons, the intensity of the light emitted will build up quickly. This cascade process in which photons emitted from excited chromium ions cause stimulated emission from other excited ions…"

> "The ruby laser is often referred to as an example of a three-level system. More than three energy levels are actually involved but they can be put into three categories. These are; the lower level form which pumping takes place, the F levels into which the chromium ions are pumped, and the metastable levels from which stimulated emission occurs. Other types of laser operate on a four level system and , in general, the mechanism of amplification differs for different lasing materials. However, in all cases, it is necessary to set up a population inversion so that stimulated emission occurs more often than absorption."

The following rate equation analysis demonstrates the critical conditions necessary to obtain sufficient inversion:

A "PUMPED" THREE-LEVEL QUANTUM SYSTEM



Master rate equations which include "laser" and "pumping" radiation, but in which thermal radiation is neglected:

$$\frac{d}{dt}N_1 = N_3\ A_{31} + N_2\ A_{21} + (N_3 - N_1)\ B_{31}\ U(\omega_P) + (N_2 - N_1)\ B_{21}\ U(\omega_L) \quad \text{[V-1a]}$$

$$\frac{d}{dt}N_2 = N_3\ A_{32} - N_2\ A_{21} - (N_2 - N_1)\ B_{21}\ U(\omega_L) \quad \text{[V-1b]}$$

$$\frac{d}{dt} N_3 = -N_3\, A_{31} - N_3\, A_{32} - \left(N_3 - N_1\right) B_{31}\, U\left(\omega_P\right) \qquad \text{[V-1c]}$$

Of course, only two of these equations are independent, since we assume that there are a fixed number of systems N distributed over these energy levels. In what follows, we define

$$R = U\left(\omega_P\right) B_{31} \left(N_1 - N_3\right) / N$$

as the ***pumping rate*** with $N_3 = N - N_1 - N_2$.

We now study the steady state solutions of two independent rate equations -- *i.e.*

$$\begin{aligned} 0 &= N_3\, A_{32} - N_2\, A_{21} - \left(N_2 - N_1\right) B_{21}\, U\left(\omega_L\right) \\ 0 &= -N_3 \left(A_{31} + A_{32}\right) - \left(N_3 - N_1\right) B_{31}\, U\left(\omega_P\right) \approx -N_3 \left(A_{31} + A_{32}\right) + R\, N \end{aligned} \qquad \text{[V-2a]}$$

which may be written

$$\begin{aligned} N_1 \left[A_{32} - B_{21}\, U\left(\omega_L\right)\right] + N_2 \left[A_{32} + A_{21} + B_{21}\, U\left(\omega_L\right)\right] &= N\, A_{32} \\ N_1 \left[A_{31} + A_{32}\right] + N_2 \left[A_{31} + A_{32}\right] &= N \left(A_{31} + A_{32}\right) - R\, N \end{aligned} \qquad \text{[V-2b]}$$

By messy, but trivial algebra, we find

$$\begin{aligned} N_1 &= -\frac{N\left[R\left(A_{32} + A_{21} + B_{21}\, U\left(\omega_L\right)\right) - \left(A_{31} + A_{32}\right)\left(A_{21} + B_{21}\, U\left(\omega_L\right)\right)\right]}{\left[A_{31} + A_{32}\right]\left[A_{21} + 2B_{21}\, U\left(\omega_L\right)\right]} \\ N_2 &= \frac{N\left[R\left(A_{32} - B_{21}\, U\left(\omega_L\right)\right) + \left(A_{31} + A_{32}\right) B_{21}\, U\left(\omega_L\right)\right]}{\left[A_{31} + A_{32}\right]\left[A_{21} + 2B_{21}\, U\left(\omega_L\right)\right]} \end{aligned} \qquad \text{[V-3]}$$

Therefore, the all important population difference may be expressed as

$$N_2 - N_1 = \frac{N\left[R\left(2A_{32} + A_{21}\right) - A_{21}\left(A_{31} + A_{32}\right)\right]}{\left[A_{31} + A_{32}\right]\left[A_{21} + 2B_{21}\,U(\omega_L)\right]} \qquad [\text{ V-4a }]$$

or, with regrouping of terms, as

$$\begin{aligned} N_2 - N_1 &= \frac{N\left[R\left(2A_{32} + A_{21}\right) - A_{21}\left(A_{31} + A_{32}\right)\right]}{A_{21}\left[A_{31} + A_{32}\right]\left[1 + \dfrac{2B_{21}\,U(\omega_L)}{A_{21}}\right]} \\ &= \frac{N\left[R\left(2A_{32} + A_{21}\right) - A_{21}\left(A_{31} + A_{32}\right)\right]}{A_{21}\left[A_{31} + A_{32}\right]\left[1 + \dfrac{I(z,\omega_L)}{I_{crit}}\right]} \end{aligned} \qquad [\text{ V-4 b}]$$

where $I_{crit} = (c/n)\left(A_{21}/2B_{21}\right)$. Since the spatial change in the Poynting vector associated with a beam propagating through the laser medium is equal to the rate of energy density lost (or gained) by the beam, **you can show** that

$$\frac{1}{I(z,\omega)}\frac{d}{dz}I(z,\omega) = -\left(N_1 - N_2\right)(c/n)\left(\hbar\,\omega\,B_{21}\right)\left(F(\omega)/V\right) \qquad [\text{ V-5 }]$$

where $F(\omega)$ is the ***line shape*** function of the $E_2 - E_1$ absorption line. Using Equation [V-4b], we may write

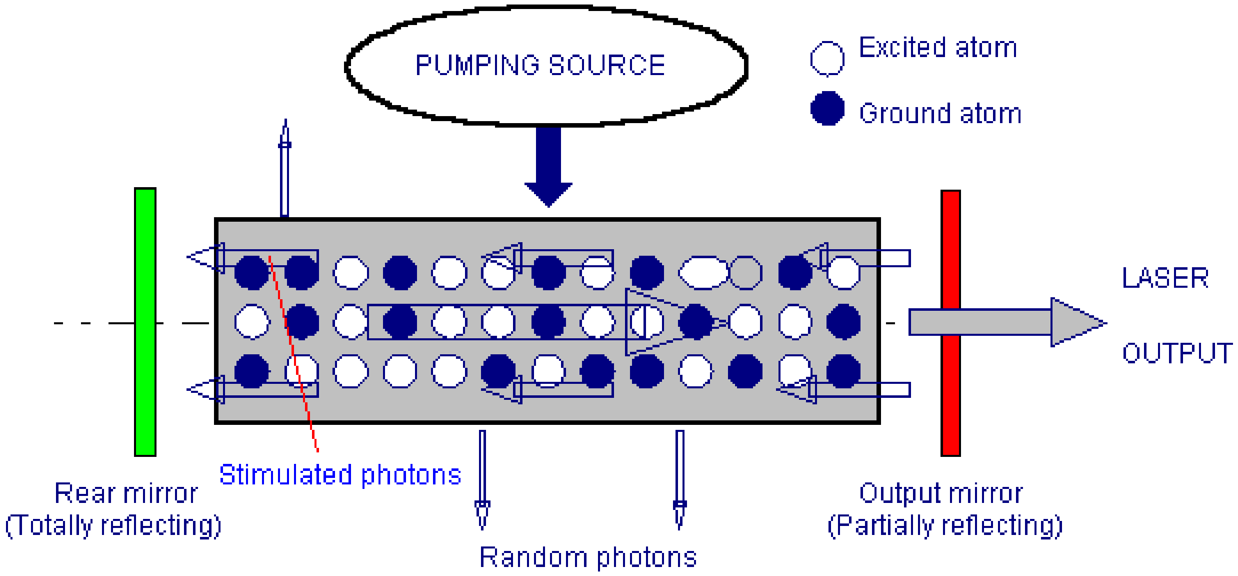
$$\frac{1}{I(z,\omega)}\frac{d}{dz}I(z,\omega) = \frac{R\left(2A_{32} + A_{21}\right) - A_{21}\left(A_{31} + A_{32}\right)}{A_{21}\left[A_{31} + A_{32}\right]\left[1 + \dfrac{I(z,\omega)}{I_{crit}}\right]}\left(\frac{c}{n}\right)\frac{N\,\hbar\,\omega\,B_{21}\,F(\omega)}{V} \qquad [\text{ V-6 }]$$

Therefore, we may write

$$\frac{1}{I(z,\omega)}\left[1+\frac{I(z,\omega)}{I_{crit}}\right]\frac{d}{dz}I(z,\omega) = \mathcal{G} \qquad [\text{ V-7a }]$$

where we identify the ***gain of the medium*** as

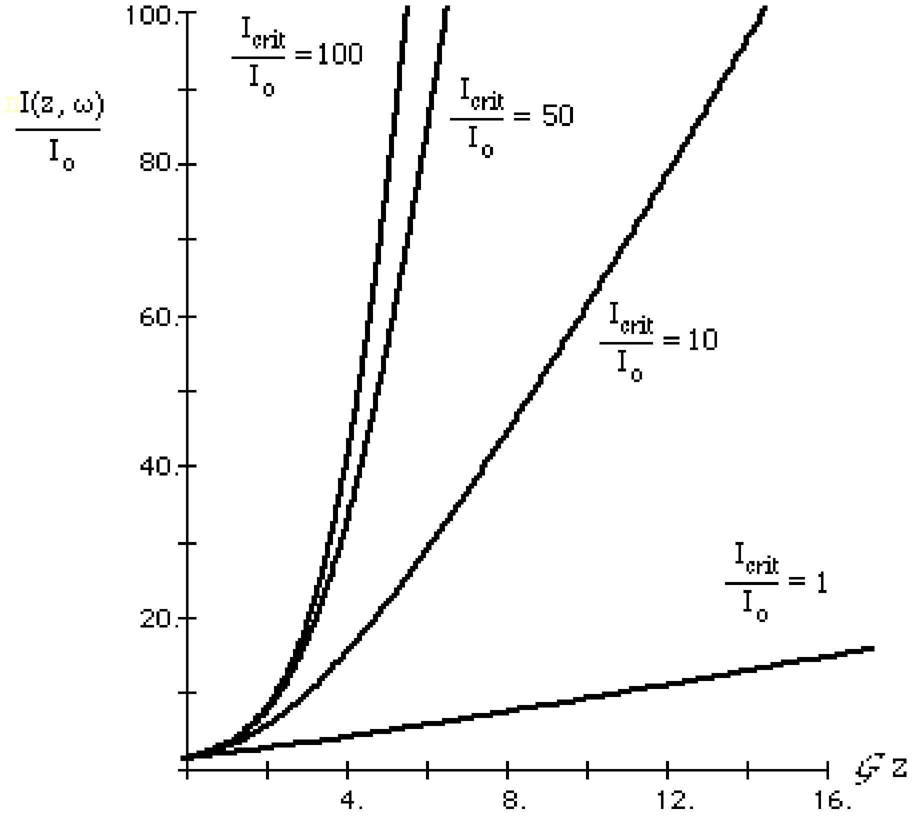
$$\mathcal{G} = \frac{R\left(2A_{32}+A_{21}\right)-A_{21}\left(A_{31}+A_{32}\right)}{A_{21}\left[A_{31}+A_{32}\right]}\left(\frac{c}{n}\right)\frac{N\,\hbar\omega\,B_{21}\,F(\omega)}{V} \qquad [\text{ V-7b }]$$



A LASER SYSTEM

Thus, we have a interesting and useful model for the "gain behavior" (growth in intensity with position) of a beam as a function of initial intensity -- *viz*.

$$\ln\left(\frac{I(z,\omega)}{I_o}\right)+\frac{I(z,\omega)-I_o}{I_{crit}} = \mathcal{G}\,z \qquad [\text{ V-8 }]$$



For $I_o \ll I_{crit}$ $\quad I(z,\omega) = I_o \exp(\mathcal{G}\, z)$ "Inverse Beer's Law"

For $I_o \gg I_{crit}$ $\quad I(z,\omega) = I_o + I_{crit}\, \mathcal{G}\, z$ Linear regime

Now consider elementary laser operation with feedback provided by two mirrors separated by a distance L. We first calculate the famous formula for ***threshold gain*** from the following consistency condition:

$$I_o \exp(-\alpha\, 2L) \exp(\mathcal{G}_{th}\, 2L)\, r_1\, r_2 = I_o \qquad \text{[V-9a]}$$

or

$$\boxed{\mathcal{G}_{th} = \alpha + \frac{1}{2L} \ln\left(\frac{1}{r_1\, r_2}\right)} \qquad \text{[V-9b]}$$

where α is included to account for residual cavity losses and the r_i 's are the respective reflectivity's of the two mirrors. At high power -- *i.e.* at powers well above threshold -- the consistency condition becomes (neglecting residual absorption)

$$I_o \approx r_1\, r_2 \left[I_o + I_{crit}\, \mathcal{G}\, 2L\right] \qquad \text{[V-10a]}$$

or

$$I_o \approx 2 I_{crit}\, \mathcal{G}\, L \left[\frac{1}{r_1\, r_2} - 1\right]^{-1} \qquad \text{[V-10b]}$$

From Equation [V-7b] we may write

$$\frac{\mathcal{G}}{\mathcal{G}_{th}} = \frac{R - R_o}{R_{th} - R_o} \qquad \text{[V-11a]}$$

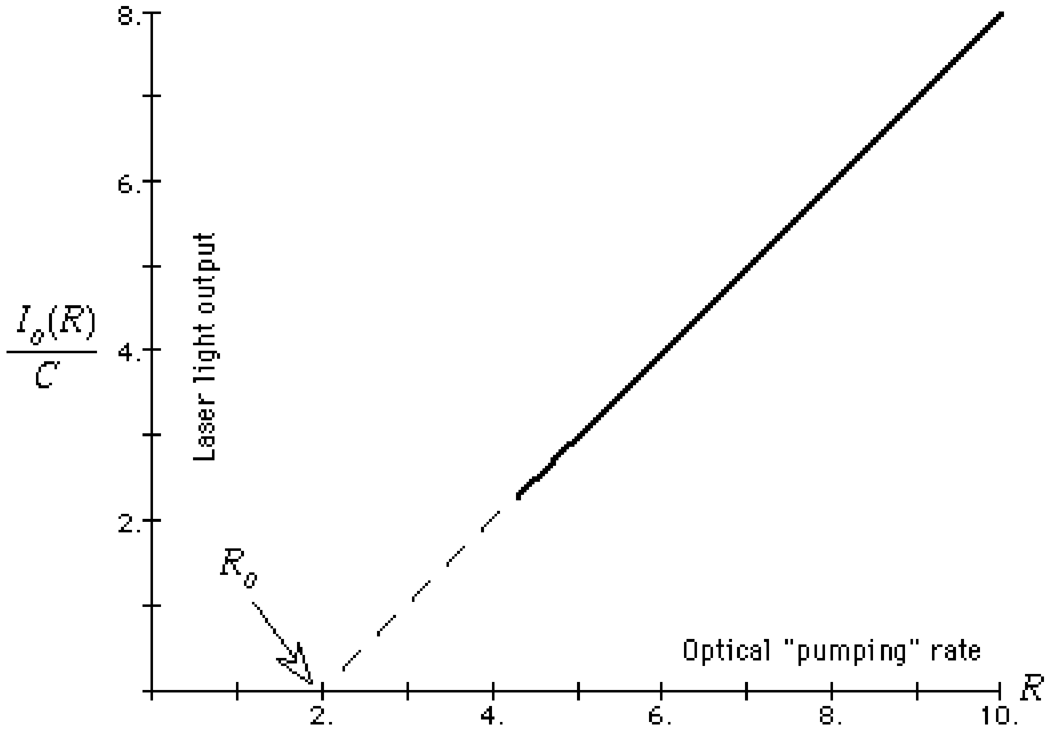
where $R_o = \dfrac{A_{21}\left(A_{31} + A_{32}\right)}{\left(2A_{32} + A_{21}\right)}$ and $R_{th} - R_o = \mathcal{G}_{th}\, \dfrac{A_{21}\left[A_{31} + A_{32}\right]}{\left(2A_{32} + A_{21}\right)} \left(\dfrac{n}{c}\right) \dfrac{V}{N\, \hbar\omega\, B_{21}\, F(\omega)}$ [V-11b]

Therefore Equation [V-10b] becomes

$$I_o(R) \approx \frac{2 I_{crit}\, \mathcal{G}_{th}\, L}{\left[\frac{1}{r_1\, r_2} - 1\right]} \left\{\frac{R - R_o}{R_{th} - R_o}\right\} = \left\{\frac{2 I_{crit}\, \mathcal{G}_{th}\, L}{\left[\frac{1}{r_1\, r_2} - 1\right]\left[R_{th} - R_o\right]}\right\} (R - R_o) = C\, (R - R_o)\ . \qquad \text{[V-12]}$$

This result, which is plotted below, is archetypal of the threshold behavior all lasers.

LASER LIGHT OUTPUT "WELL ABOVE" THRESHOLD

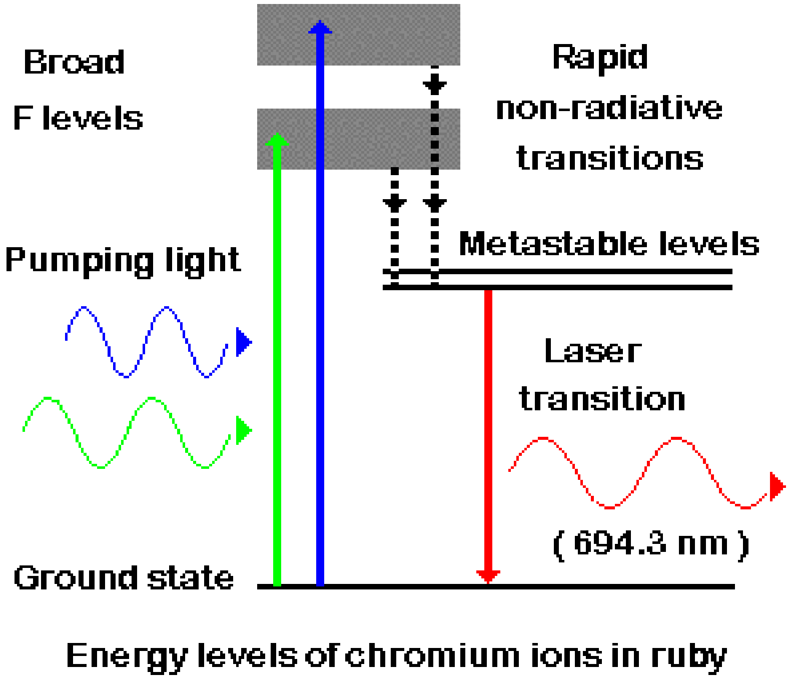


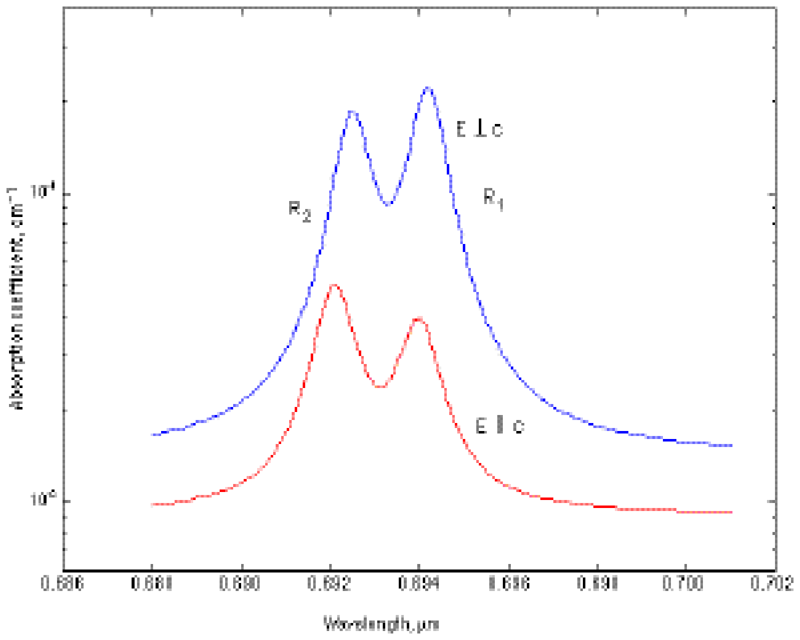
Appendix: Common Lasers

Pink Ruby Laser Laser: the first pulsed laser

Energy Levels of Cr^{3+} in a Trigonal Crystal Field

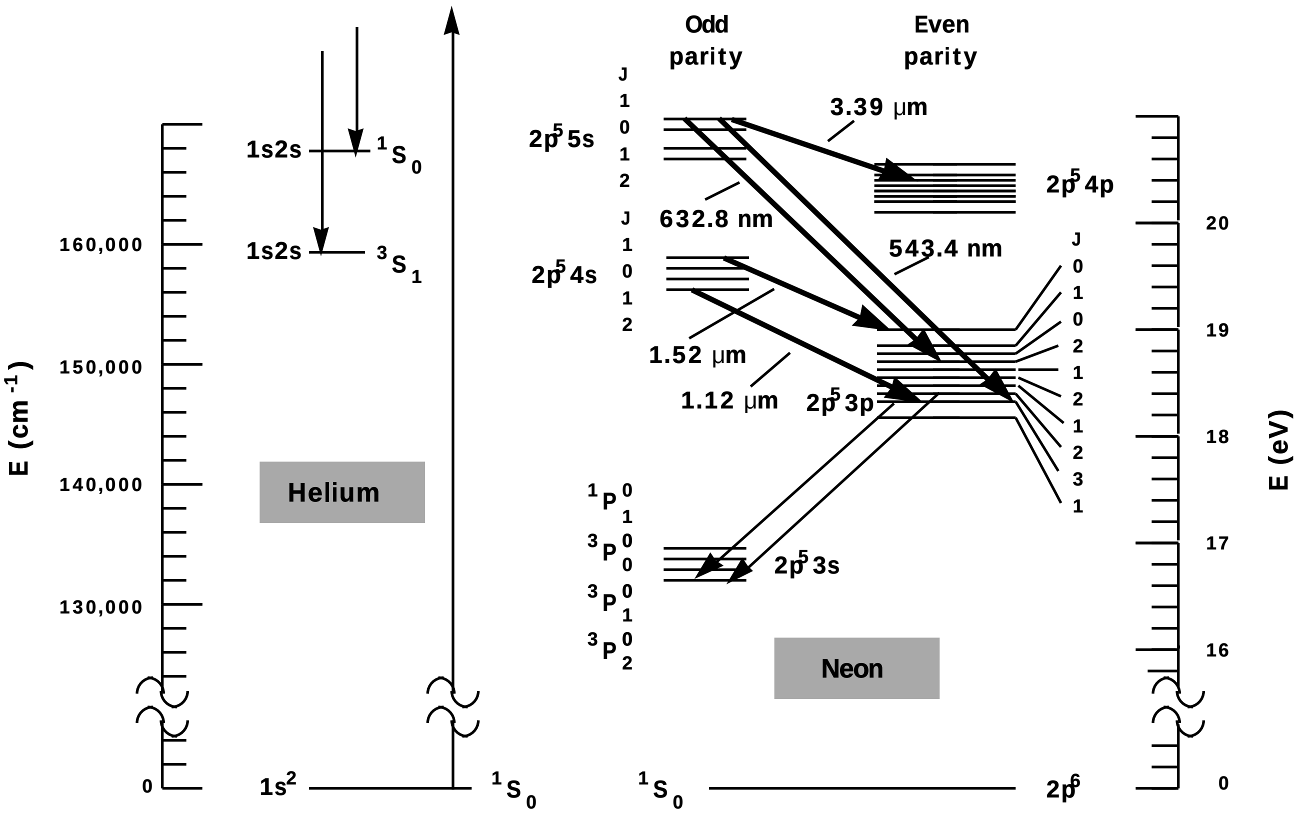
Absorption Spectrum of R1 and R2 of Cr^{3+} in Ruby





Absorption Spectrum of R1 and R2 of Cr^{3+} in Rub

HeNe Neutral Gas Laser: the first continuous laser



Mechanism: A dc or rf electrical discharge is established in a gas mixture of helium (≈ 90%) and neon (≈10%). The helium atoms are excited to long-lived metastable states by a variety of processes involving energetic electron impact. The excitation energy of the helium metastables is transfered to neon atoms by resonance scattering to yield **laser-active** neon metastables.

Particularly strong emissions

$1s^22s^22p^55s$: $^1P_1$ → $1s^22s^22p^53p$: $^3S_1$ $\lambda_{vacuo} = 0.5435$ μm; $\lambda_{air} = 0.5433$ μm

$1s^22s^22p^55s$: $^1P_1$ → $1s^22s^22p^53p$: $^3P_2$ $\lambda_{vacuo} = 0.6330$ μm; $\lambda_{air} = 0.6328$ μm

$1s^2 2s^2 2p^5 4s$: ${}^1P_1$ → $1s^2 2s^2 2p^5 3p$: ${}^3P_2$ $\lambda_{vacuo} = 1.1526\ \mu m$; $\lambda_{air} = 1.1523\ \mu m$

$1s^2 2s^2 2p^5 4s$: ${}^1P_1$ → $1s^2 2s^2 2p^5 3p$: ${}^1S_0$ $\lambda_{vacuo} = 1.5235\ \mu m$; $\lambda_{air} = 1.5231\ \mu m$

$1s^2 2s^2 2p^5 5s$: ${}^1P_1$ → $1s^2 2s^2 2p^5 4p$: ${}^3P_2$ $\lambda_{vacuo} = 3.3922\ \mu m$; $\lambda_{air} = 3.3913\ \mu m$

Rare Earth Ion Solid State Lasers:

Neutral atom electronic configurations:

noble gas core xeon: $[Xe] \equiv 1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^6 4d^{10} 5s^2 5p^6$

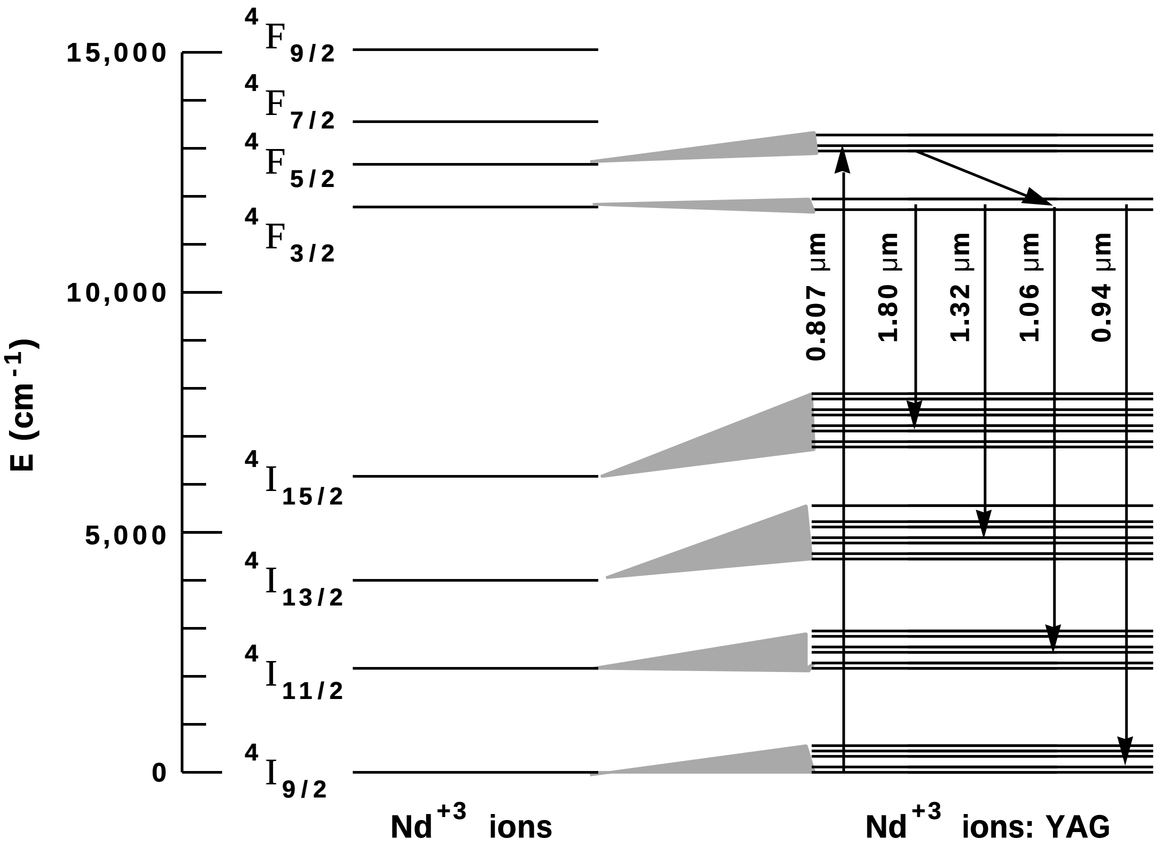
rare earth atoms: $1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^6 4d^{10} 4f^{\,n} 5s^2 5p^6 6s^2 = [Xe]\ 4f^{\,n} 6s^2$

Trivalent ion electronic configurations of common laserable rare earth ions:

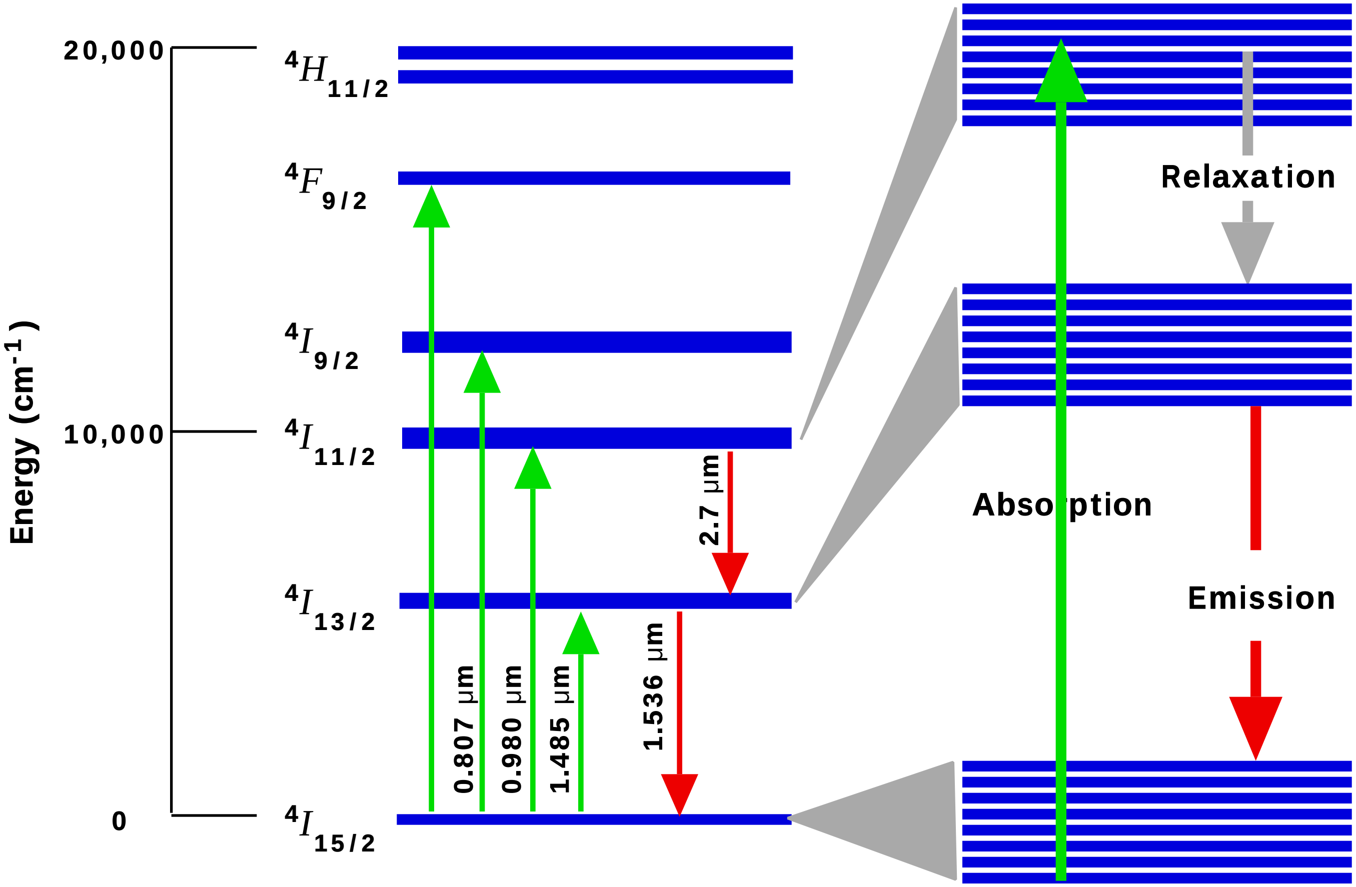
praseodymium (Pr^{+3}):	$= [Xe]\ 4f^{\,2}$	Ground state: ${}^3H_4$
neodymium (Nd^{+3}):	$= [Xe]\ 4f^{\,3}$	Ground state: ${}^4I_{9/2}$
europium (Eu^{+3}):	$= [Xe]\ 4f^{\,6}$	Ground state:
holmium (Ho^{+3}):	$= [Xe]\ 4f^{\,10}$	Ground state:
erbium (Er^{+3}):	$= [Xe]\ 4f^{\,11}$	Ground state: ${}^4I_{15/2}$
thulium (Tm^{+3}):	$= [Xe]\ 4f^{\,12}$	Ground state: ${}^3H_6$
ytterbium (Yb^{+3}):	$= [Xe]\ 4f^{\,13}$	

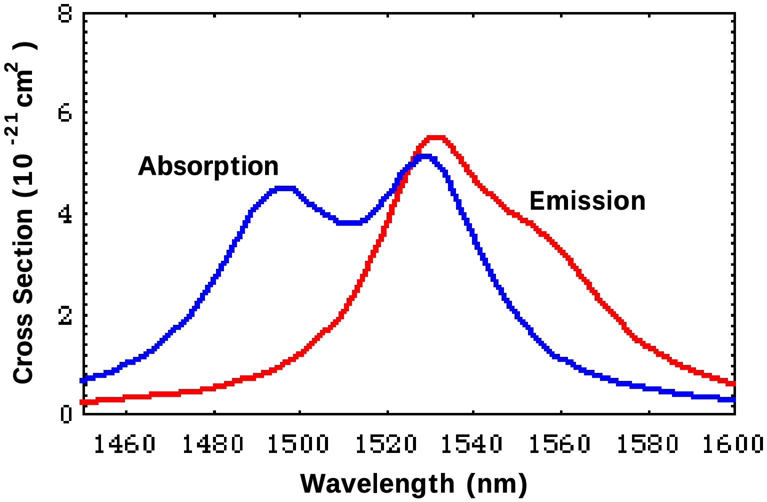
Important characteristic of rare earth ions: Outer 5*s* and 5*p* shells effectively shield inner 4*f* electrons from significant interaction with the local crystalline field associated with the charges on neighboring ions in solid state configurations.

1. YIG Laser: Nd^{+3} randomly distributed on Y^{+3} sites in $Y_3Al_5O_{12}$ (garnet)



2. EDFA (Erbium Doped Fiber Amplifier): Er^{+3} randomly distributed in glass


Energy (cm^{-1})
20,000
10,000
0
$^4H_{11/2}$
$^4F_{9/2}$
$^4I_{9/2}$
$^4I_{11/2}$
$^4I_{13/2}$
$^4I_{15/2}$
0.807 µm
0.980 µm
1.485 µm
1.536 µm
2.7 µm
Relaxation
Absorption
Emission

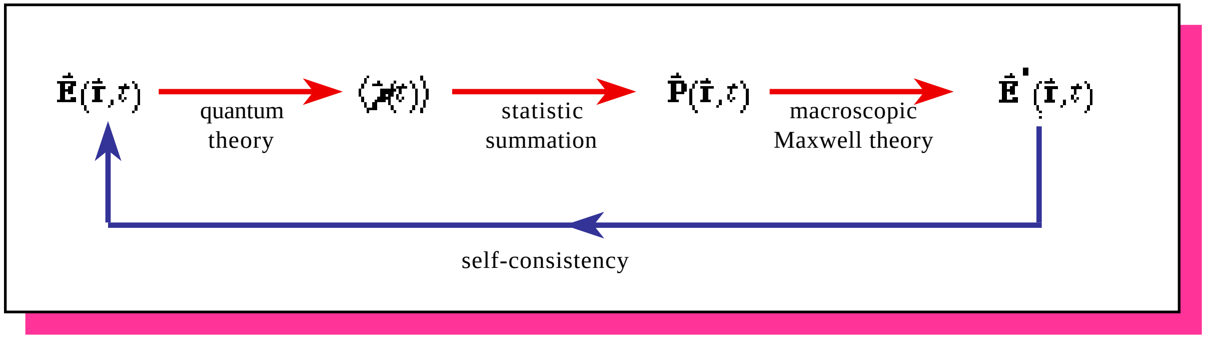


The absortion and emmission cross sections for the $^4I_{15/2} \rightarrow {}^4I_{13/2}$ transition in Er^{+3}.

VI. SEMICLASSICAL LASER THEORY [23]

LASER SELF-CONSISTENCY EQUATIONS

Lamb's theory of laser operation provides a very powerful means for interpreting and predicting complex time dependent behavior without invoking all the intricacy of a full quantum mechanical theory. It is semiclassical, self-consistent theory in the following sense:



Suppose that the field in the laser is represented in the form

$$E(z,t) = \frac{1}{2}\sum_{n} \mathcal{E}_n(t)\, \exp\left[-i\left(\omega_n t + \varphi_n(t)\right)\right] U_n(z) + c.c. \qquad \text{[VI-1]}$$

where, for example,

in a simple a cavity laser $U_n(z) \quad \sin k_n z = \sin\left(\frac{n\,\pi}{L} z\right)$ **standing wave** [VI-2a]

and in a ring laser $U_n(z) \quad \exp\left(\, i\, k_n\, z\right) = \exp\left(i\,\frac{n\,2\pi}{L} z\right)$ **traveling wave** [VI-2b]

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[23] An adaptation or interpretation of Lamb's "semiclassical" or "self-consistent" laser theory as first presented in *Phys. Rev.* **134**, A1429 (1964) and refined in countless other treatments. In these lecture notes we drawn extensively on M. Sargent III, M. O. Scully and W. E. Lamb, Jr., *Laser Physics*, Addison-Wesley (1974) and P. Meystre and M. Sargent III, *Elements of Quantum Optics*, Springer-Verlag (1992).

With this representation for the field the **induced polarization** can be expressed as

$$P(z,t) = \frac{1}{2}\sum_{n} \mathcal{P}_n(t) \exp\left[-i\left(\omega_n t + \varphi_n(t)\right)\right] U_n(z) + \text{c.c.} \qquad [\text{ VI-3 }]$$

where $\mathcal{P}_n(t)$ is the complex, slowly varying component of the polarization of the nth mode. If we take the wave equation in the form[24]

$$- \;{}^{2}\vec{\mathbf{E}} + \mu_o \frac{\partial \vec{\mathbf{J}}}{\partial t} + \frac{1}{c^2}\frac{\partial^2 \vec{\mathbf{E}}}{\partial t^2} = -\mu_o \frac{\partial^2 \vec{\mathbf{P}}}{\partial t^2} \qquad [\text{ VI-4 }]$$

where second term is included as a means to account for cavity losses. From Equation [VI-1] assuming that $\mathcal{E}_n$, $\mathcal{P}_n$, and $\dot{\varphi}_n$ are slowly varying functions of time[25]

$$\begin{aligned} - \;{}^{2}\vec{\mathbf{E}} \quad - \frac{\partial^2 E(z,t)}{\partial z^2} &= -\frac{1}{2}\sum_{n} \mathcal{E}_n \exp\left[-i\left(\omega_n t + \varphi_n\right)\right] \frac{\partial^2 U_n}{\partial z^2} + c.c. \\ &= \frac{1}{2c^2}\sum_{n} \Omega_n^2\, \mathcal{E}_n \exp\left[-i\left(\omega_n wt + \varphi_n\right)\right] U_n + c.c. \end{aligned} \qquad [\text{ VI-5a }]$$

where $\Omega_n = c\,k_n$ are the eigenfrequencies of the ***cold resonator*** eigenmodes.

$$\frac{\partial \vec{\mathbf{E}}}{\partial t} \quad \frac{\partial E(z,t)}{\partial t} = \frac{1}{2}\sum_{n}\left\{\dot{\mathcal{E}}_n - i\,\mathcal{E}_n\left(\omega_n + \dot{\varphi}_n\right)\right\} \exp\left[-i\left(\omega_n t + \varphi_n\right)\right] U_n(z) + c.c. \qquad [\text{ VI-5b }]$$

---

[24] In this formulation we are assuming that $\overline{}\;\vec{\mathbf{P}} \approx 0$.

[25] The so called ***slowly-varying amplitude and phase approximation*** (SVAP) is used extensively in treating problems in laser dynamics. In the SVAP approximation it is assumed that

$$\left\{\frac{\partial}{\partial z}\ln\left|\mathcal{E}_n\right|;\ \frac{\partial}{\partial z}\ln\left|\mathcal{P}_n\right|;\ \frac{\partial}{\partial z}\left|\varphi_n\right|\right\} << k_n \text{ and } \left\{\frac{\partial}{\partial t}\ln\left|\mathcal{E}_n\right|;\ \frac{\partial}{\partial t}\ln\left|\mathcal{P}_n\right|;\ \frac{\partial}{\partial t}\left|\varphi_n\right|\right\} << \Omega_n$$

$$\mu_o \frac{\partial \vec{\mathbf{J}}}{\partial t} \quad \mu_o \frac{\partial J(z,t)}{\partial t} \approx \frac{\mu_o \sigma}{2}\sum_n \left\{-i\,\mathcal{E}_n\,\omega_n\right\}\exp\left[-i\left(\omega_n t+\varphi_n\right)\right]U_n(z)+c.c. \qquad \text{[VI-5c]}$$

$$\frac{\partial^2 \vec{\mathbf{E}}}{\partial t^2} \quad \frac{\partial^2 E\,w(z,t)}{\partial t^2} = \frac{1}{2}\sum_n \left\{\ddot{\mathcal{E}}_n - i\,2\,\dot{\mathcal{E}}_n\left(\omega_n+\dot{\varphi}_n\right)+\mathcal{E}_n\left[-\left(\omega_n+\dot{\varphi}_n\right)^2 - i\,\ddot{\varphi}_n\right]\right\}\exp\left[-i\left(\omega_n t+\varphi_n\right)\right]U_n(z)+c.c. \qquad \text{II-5d]}$$

which reduces in the **SVAP** approximation to

$$\frac{\partial^2 \vec{\mathbf{E}}}{\partial t^2} \quad \frac{\partial^2 E(z,t)}{\partial t^2} \approx \frac{1}{2}\sum_n \left\{-i\,2\,\dot{\mathcal{E}}_n\,\omega_n+\mathcal{E}_n\left[-\omega_n^2-2\omega_n\,\dot{\varphi}_n\right]\right\}\exp\left[-i\left(\omega_n\,wt+\varphi_n\right)\right]U_{nw}(z)+c.c. \qquad \text{[VI-5d']}$$

and from Equation [VI-3]

$$\begin{aligned}\frac{\partial^2 \vec{\mathbf{P}}}{\partial t^2} \quad \frac{\partial^2 P(z,t)}{\partial t^2} &= \frac{1}{2}\sum_n \left\{\ddot{\mathcal{P}}_n - i\,2\,\dot{\mathcal{P}}_n\left(\omega_n+\dot{\varphi}_n\right)+\mathcal{P}_n\left[-\left(\omega_n+\dot{\varphi}_n\right)^2 - i\,\ddot{\varphi}_n\right]\right\}\exp\left[-i\left(\omega_n t+\varphi_n\right)\right]U_n(z)+c.c. \\ &\approx \frac{1}{2}\sum_n \left\{-\omega_n^2\,\mathcal{P}_n\right\}\exp\left[-i\left(\omega_n t+\varphi_n\right)\right]U_n(z)+c.c.\end{aligned} \qquad \text{[VI-6]}$$

If these representations are to be valid when substituted into Equation [VI-4], the following equations must hold:

$$\Omega_n^2\,\mathcal{E}_n - i\left(\frac{\sigma}{\varepsilon_o}\right)\omega_n\,\mathcal{E}_n - i\,2\omega_n\,\dot{\mathcal{E}}_n - \omega_n^2\,\mathcal{E}_n - 2\omega_n\,\dot{\varphi}_n\,\mathcal{E}_n \approx \frac{1}{\varepsilon_o}\,\omega_n^2\,\mathcal{P}_n \qquad \text{[VI-7]}$$

We adjust the ***fictional*** conductivity to account for the loss of energy or time decay of the given mode -- *viz.* we take $\sigma = \varepsilon_o\,\omega_n / Q_n$ where Q_n is the "Q" of the mode. Equating real and imaginary parts of Equation [VI-7] we obtain Lamb's **master laser self-consistency equations** -- *viz.*

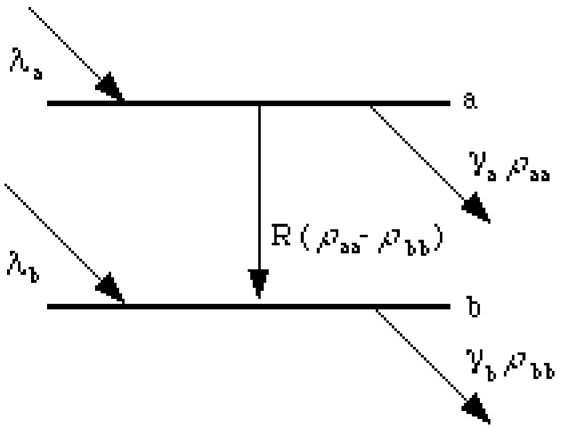
$$\dot{\mathcal{E}}_n + \left(\frac{\omega_n}{2Q_n}\right)\mathcal{E}_n \approx -\frac{\omega_n}{2\varepsilon_o}\,\mathcal{I}m\left\{\,\mathcal{P}_n\right\} \qquad \text{[VI-8a]}$$

$$\Omega_n^2\,\mathcal{E}_n - \omega_n^2\,\mathcal{E}_n - 2\omega_n\,\dot{\varphi}_n\,\mathcal{E}_n \approx \frac{1}{\varepsilon_o}\omega_n^2\,\mathcal{R}e\left\{\,\mathcal{P}_n\right\} \qquad \text{[VI-8b]}$$

$$\text{or} \qquad \omega_n + \dot{\varphi}_n \approx \Omega_n - \frac{\omega_n}{2\varepsilon_o}\,\mathcal{R}e\left\{\,\mathcal{P}_n\right\}\Big/\mathcal{E}_n \qquad \text{[VI-8b']}$$

POLARIZATION OF THE MEDIUM:

The simplest application of Lamb's theory assumes that the laser medium consists of an ensemble of two-level atoms with a single well-defined transition $\hbar\omega_{ab}$ which is **homogeneously broaden**. Possible "pump" and "decay" processes are indicated schematically in the following figure:



The model describes, in effect, a so called ***four-level*** **homogeneously broaden laser** where both levels relaxed to some presumed *ground state* and are "pumped" via higher energy excited states. $\rho\left(s,z,t',t\right)$ is defined as the density matrix operator at a place z and a time t which is associated with atoms excited to the state s (where $s = a$ or b) at a time t' and $\lambda_s\left(z,t'\right)$ as the rate at which atoms are excited to the state s (per atom, per second per unit volume). The macroscopic polarization is, thus, given by

$$P\left(z,t\right) = \sum_{\mathrm{sw}} \int_{-\infty}^{t} d\,t'\, \lambda_{\mathrm{s}}\left(z,t'\right) \left\langle e\, \mathcal{D}isp \right\rangle$$

$$= \sum_{\mathrm{s}} \int_{-\infty}^{t} d\,t'\, \lambda_{\mathrm{s}}\left(z,t'\right) \rho_{\mathrm{ab}}\left(s,z,t',t\right) + c.c. \qquad \text{[VI-9]}$$

If we project this expression on to the modes of the laser cavity and compare with Equation [VI-3], we see that

$$\mathcal{P}_{\mathrm{n}}\left(t\right) = 2 \quad \exp\left[\, i\left(\omega_{\mathrm{n}}\, t + \varphi_{\mathrm{n}}\left(t\right)\right)\right] \frac{1}{\mathcal{M}_{\mathrm{n}}} \int_0^L dz U_{\mathrm{n}}\left(z\right) \sum_{\mathrm{s}} \int_{-\infty}^{t} d\,t'\, \lambda_{\mathrm{s}}\left(z,t'\right) \rho_{\mathrm{ab}}\left(s,z,t',t\right) \qquad \text{[VI-10a]}$$

where $\mathcal{M}_{\mathrm{n}} = \int_0^L dz \left| U_{\mathrm{n}}\left(z\right) \right|^2$ is the mode normalization factor. To facilitated the integration of this equation, we re-write it in terms of the **population matrix operator** $\bar{\rho}\left(z,t\right)$ -- *viz.*

$$\mathcal{P}_{\mathrm{n}}\left(t\right) = 2 \quad \exp\left[\, i\left(\omega_{\mathrm{n}}\, t + \varphi_{\mathrm{n}}\left(t\right)\right)\right] \frac{1}{\mathcal{M}_{\mathrm{n}}} \int_0^L dz\, U_{\mathrm{n}}\left(z\right) \bar{\rho}_{\mathrm{ab}}\left(z,t\right) \qquad \text{[VI-10b]}$$

where

$$\bar{\rho}\left(z,t\right) \equiv \sum_{\mathrm{s}} \int_{-\infty}^{t} d\,t'\, \lambda_{\mathrm{s}}\left(z,t'\right) \rho\left(s,z,t',t\right) \, . \qquad \text{[VI-11]}$$

The equation of motion of this population matrix is found by differentiating its defining equation -- *i.e.*

$$\frac{d\bar{\rho}\left(z,t\right)}{d\,t} = \sum_{s} \lambda_{s}\left(z,t\right)\rho\left(s,z,t,t\right) + \sum_{s}\int_{-\infty}^{t} d\,t'\,\lambda_{s}\left(z,t'\right)\dot{\rho}\left(s,z,t',t\right) . \qquad \text{[VI-12]}$$

By definition $\rho_{\alpha\beta}\left(s,z,t,t\right) = \delta_{\alpha\,s}\,\delta_{\beta\,s}$, so that first term on the right-hand-side is replaced by the matrix

$$\begin{bmatrix} \lambda_{a}\left(z,t\right) & 0 \\ 0 & \lambda_{b}\left(z,t\right) \end{bmatrix}$$

If we interpret the $\dot{\rho}\left(s,z,t',t\right)$ components of the second term on the right-hand-side of Equation [VI-12], as the time derivatives of the ***pure state*** density matrix components given by Equations [III-14a] and [III-14b] of this set of lectures notes,[26] the component equations of motion for the population matrix $\bar{\rho}\left(z,t\right)$ become

$$\dot{\bar{\rho}}_{aa} = \lambda_{a} - \gamma_{a}\,\bar{\rho}_{aa} - \left[i\,\hbar^{-1}\mathcal{V}_{ab}\,\bar{\rho}_{ab} + c.c.\right] \qquad \text{[VI-13a]}$$

$$\dot{\bar{\rho}}_{bb} = \lambda_{b} - \gamma_{b}\,\bar{\rho}_{bb} + \left[i\,\hbar^{-1}\mathcal{V}_{ab}\,\bar{\rho}_{ab} + c.c.\right] \qquad \text{[VI-13b]}$$

$$\dot{\bar{\rho}}_{ab} = -\left(i\,\omega_{ab} + \gamma_{ab}\right)\bar{\rho}_{ab} + i\,\hbar^{-1}\mathcal{V}_{ab}\left[\bar{\rho}_{aa} - \bar{\rho}_{bb}\right] \qquad \text{[VI-13c]}$$

Formal integration of Equation [VI-13c] yields

---

[26] Including damping, Equations [III-16a] and [III-16b] of this set of lecture notes would have the form

$$\dot{\rho}_{aa} = -\gamma_{a}\,\rho_{aa} - \left[i\,\hbar^{-1}\mathcal{V}_{ab}\,\rho_{ab} + c.c.\right]$$

$$\dot{\rho}_{bb} = -\gamma_{b}\,\rho_{bb} + \left[i\,\hbar^{-1}\mathcal{V}_{ab}\,\rho_{ab} + c.c.\right]$$

$$\dot{\rho}_{ab} = -\left(i\,\omega_{ab} + \gamma_{ab}\right)\rho_{ab} + i\,\hbar^{-1}\mathcal{V}_{ab}\left[\rho_{aa} - \rho_{bb}\right]$$

$$\rho_{ab} = i\,\hbar^{-1} \int_{-\infty}^{t} dt' \exp w\left[-\left(i\,\omega_{ab} + \gamma_{ab}\right)\left(t - t'\right)\right] \mathcal{V}_{ab}\left(z, t'\right) \left[\rho_{aa}\left(z, t'\right) - \rho_{bb}\left(z, t'\right)\right] \quad \text{[VI-14]}$$

SINGLE MODE OPERATION:

For a **single-mode standing wave field** in the **rotating-wave approximation**

$$\mathcal{V}_{ab}\left(z, t'\right) = -\frac{1}{2} \quad \mathcal{E}_{n}(t) \exp\left[-i\left(\omega_{n} t' + \varphi_{n}\left(t'\right)\right)\right] U_{n}(z) \qquad \text{[VI-15]}$$

so that

$$\begin{aligned} \rho_{ab}(z, t) &= -\frac{1}{2}\, i \quad \hbar^{-1} \exp\left[-i\,\omega_{n} t\right] U_{n}(z) \int_{-\infty}^{t} dt' \exp\left\{-\left[i\left(\omega_{ab} - \omega_{n}\right) + \gamma_{ab}\right]\left(t - t'\right) - i\,\varphi_{n}\left(t'\right)\right\} \mathcal{E}_{n}\left(t'\right) \left[\rho_{aa}\left(z, t'\right) - \rho_{bb}\left(z, t'\right)\right] \\ &= -\frac{1}{2}\, i \quad \hbar^{-1} \exp\left[-i\,\omega_{n} t\right] U_{n}(z) \int_{0}^{+\infty} d\tau \exp\left\{-\left[i\left(\omega_{ab} - \omega_{n}\right) + \gamma_{ab}\right]\tau - i\,\varphi_{n}(t - \tau)\right\} \mathcal{E}_{n}(t - \tau) \left[\rho_{aa}(z, t - \tau) - \rho_{bb}(z, t - \tau)\right] \end{aligned} \quad \text{[VI-16]}$$

The integration can be done simply, if the changes in $\mathcal{E}_{n}$, φ_{n}, and $\rho_{aa} - \rho_{bb}$ are negligible in a time γ_{ab}^{-1} so that

$$\begin{aligned} \rho_{ab}(z, t) &= -\frac{1}{2}\, i \quad \hbar^{-1} \mathcal{E}_{n}(t) \exp\left[-i\left(\omega_{n}\, t + \varphi_{n}(t)\right)\right] U_{n}(z) \left[\rho_{aa}(z, t) - \rho_{bb}(z, t)\right] \left[i\left(\omega_{ab} - \omega_{n}\right) + \gamma_{ab}\right]^{-1} \\ &= -\frac{1}{2}\, i \quad \hbar^{-1} \mathcal{E}_{n}(t) \exp\left[-i\left(\omega_{n}\, t + \varphi_{n}(t)\right)\right] U_{n}(z) \left[\rho_{aa}(z, t) - \rho_{bb}(z, t)\right] \mathcal{D}\left(\omega_{ab} - \omega_{n}; \gamma_{ab}\right) \end{aligned} \quad \text{[VI-17]}$$

where $\mathcal{D}(u - v; w) \equiv \left[i\,(u - v) + w\right]^{-1}$ represents the so called **complex Lorentzian denominator**. Substituting this result into Equations [VI-13a] and [VI-13b] we find the previously discussed **rate equation approximation** for the population components -- *viz*.

$$\begin{aligned}\dot{\rho}_{aa} &= \lambda_a - \gamma_a\,\rho_{aa} - R\left(\rho_{aa} - \rho_{bb}\right)\\ \dot{\rho}_{bb} &= \lambda_b - \gamma_b\,\rho_{bb} + R\left(\rho_{aa} - \rho_{bb}\right)\end{aligned} \qquad [\text{ VI-18 }]$$

with the **rate constant** is given by

$$\begin{aligned}R &= \frac{1}{2}\left(\frac{\;}{\hbar}\right)^{2}\mathcal{E}_n^{\,2}(t)\,|\,U_n(z)\,|^{2}\,\gamma_{ab}\left[\left(\omega_{ab}-\omega_n\right)^{2}+\gamma_{ab}^{2}\right]^{-1}\\ &= \frac{1}{2}\left(\frac{\;}{\hbar}\right)^{2}\mathcal{E}_n^{\,2}(t)\,|\,U_n(z)\,|^{2}\,\gamma_{ab}^{-1}\,\mathcal{L}\left(\omega_{ab}-\omega_n;\gamma_{ab}\right)\end{aligned} \qquad [\text{ VI-19 }]$$

where $\mathcal{L}(u-v;\,w) \equiv w^{2}\left[(u-v)^{2}+w^{2}\right]^{-1}$ represents the **dimensionless Lorentzian function**.

For steady state -- *i.e.* $\dot{\rho}_{aa} = \dot{\rho}_{bb} = 0$

$$\rho_{aa} - \rho_{bb} = \frac{\lambda_a\,\gamma_a^{-1} - \lambda_b\,\gamma_b^{-1}}{1 + R(z)\Big/\left(\dfrac{\gamma_a\,\gamma_b}{\gamma_a+\gamma_b}\right)} = \frac{N(z)}{1 + R(z)/R_S} \qquad [\text{ VI-20 }]$$

where $N(z)$ is the unsaturated population difference and $R_S = \gamma_a\,\gamma_b\left(\gamma_a+\gamma_b\right)^{-1}$. Combining Equations [VI-10b], [VI-17] and [VI-20] we obtain

$$\mathcal{P}_n(t) = -\;\;{}^{2}\,\hbar^{-1}\mathcal{E}_n\;\mathcal{D}\left(\omega_{ab}-\omega_n;\gamma_{ab}\right)\frac{1}{\mathcal{M}_n}\int_0^L dz\,\frac{N(z)\,|U_n(z)|^{2}}{1+R(z)/R_S} \qquad [\text{ VI-21 }]$$

Although this result **can be integrated exactly**,[27] there is more insight to be gained by expanding the ***saturation denominator*** in powers of $R(z)/R_S$ -- *viz.*

$$\mathcal{P}_n(t) = -\ {}^{2}\,\hbar^{-1}\mathcal{E}_n\ \mathcal{D}(\omega_{ab}-\omega_n;\gamma_{ab})\,\frac{1}{\mathcal{M}_n}\int_0^L dz\ N(z)\,|U_n(z)|^2\sum_{k=0}^{\infty}\left(-\frac{R(z)}{R_S}\right)^k \qquad [\text{ VI-22a }]$$

To second order in $R(z)/R_S$ we obtain

$$\mathcal{P}_n(t) \approx -\ {}^{2}\,\hbar^{-1}\mathcal{E}_n\ \mathcal{D}(\omega_{ab}-\omega_n;\gamma_{ab})\ \overline{N}\left\{1-\frac{3}{4}\,I_n\left(\frac{\gamma_a+\gamma_b}{\gamma_{ab}}\right)\mathcal{L}(\omega_{ab}-\omega_n;\gamma_{ab})\right\} \qquad [\text{ VI-22b }]$$

where

$$\overline{N}(t) \equiv \frac{1}{\mathcal{M}_n}\int_0^L dz\ N(z,t)\,|U_n(z)|^2 \qquad [\text{ VI-23a }]$$

and

$$I_n = \frac{1}{2}\,\frac{{}^{2}\,\mathcal{E}_n^{\,2}}{\hbar^2}\,\frac{1}{\gamma_a\,\gamma_b}\,. \qquad [\text{ VI-23b }]$$

is the dimensionless mode intensity.

To complete the self-consistency loop, we combine our expression for the complex mode polarization, Equation [VI-22b], and the self-consistency conditions, Equations [VI-8].

From Equation [VI-8a]

---

[27] Exactly

$$\mathcal{P}_n(t) = -\ {}^{2}\,\hbar^{-1}\mathcal{E}_n\ \mathcal{D}(\omega_{ab}-\omega_n;\gamma_{ab})\ \overline{N}\ f(w)$$

where

$$f(w) = \frac{2}{w}\left[1-(1+w)^{-1/2}\right] \quad \text{and} \quad w = I_n\left(\frac{\gamma_a+\gamma_b}{\gamma_{ab}}\right)\mathcal{L}(\omega_{ab}-\omega_n;\gamma_{ab})$$

$$\dot{\mathcal{E}}_{n} + \left(\frac{\omega_{n}}{2Q_{n}}\right)\mathcal{E}_{n} \quad -\frac{\omega_{n}}{2\varepsilon_{o}}\,\mathcal{Im}\left\{\mathcal{P}_{n}\right\}$$

$$\frac{1}{2}\omega_{n}\frac{{}^{2}\,\mathcal{E}_{n}}{\hbar\varepsilon_{o}}\left(\frac{1}{\gamma_{ab}}\right)\mathcal{L}\left(\omega_{ab}-\omega_{n};\gamma_{ab}\right)\,\overline{N}\left\{1-\frac{3}{4}\,I_{n}\left(\frac{\gamma_{a}+\gamma_{b}}{\gamma_{ab}}\right)\mathcal{L}\left(\omega_{ab}-\omega_{n};\gamma_{abw}\right)\right\}. \qquad [\text{ VI-24 }]$$

Multiplying this equation by $\frac{{}^{2}\,\mathcal{E}_{n}}{2\,\hbar\,\gamma_{b}\,\gamma_{b}}$, we obtain

$$\dot{I}_{n} \approx 2I_{n}\left\{-\left(\frac{\omega_{n}}{2Q_{n}}\right)+\frac{1}{2}\omega_{n}\frac{{}^{2}}{\hbar\varepsilon_{o}}\left(\frac{1}{\gamma_{ab}}\right)\mathcal{L}\left(\omega_{ab}-\omega_{n};\gamma_{ab}\right)\,\overline{N}\left[1-\frac{3}{4}\,I_{n}\left(\frac{\gamma_{a}+\gamma_{b}}{\gamma_{ab}}\right)\mathcal{L}\left(\omega_{ab}-\omega_{nw};\gamma_{ab}\right)\right]\right\} \qquad [\text{ VI-25a }]$$

This nonlinear equation of motion may be written to advantage as

$$\dot{I}_{n} \approx 2I_{n}\left[\alpha_{n}-\beta_{n}\,I_{n}\right] \qquad [\text{ VI-25b }]$$

where the ***linear gain*** is measured by

$$\alpha_{n} = \frac{1}{2}\omega_{n}\frac{{}^{2}}{\hbar\varepsilon_{o}}\left(\frac{1}{\gamma_{ab}}\right)\mathcal{L}\left(\omega_{ab}-\omega_{n};\gamma_{ab}\right)\,\overline{N}-\left(\frac{\omega_{n}}{2Q_{n}}\right) \qquad [\text{ VI-26a }]$$

and ***nonlinear saturation*** by

$$\beta_{n} = \frac{3}{8}\,\overline{N}\,\frac{\omega_{n}\quad{}^{2}}{\hbar\varepsilon_{o}\,\gamma_{ab}}\left(\frac{\gamma_{a}+\gamma_{b}}{\gamma_{ab}}\right)\mathcal{L}^{2}\left(\omega_{ab}-\omega_{n};\gamma_{ab}\right) \qquad [\text{ VI-26b }]$$

From Equation [VI-8b'], we see that

$$\omega_{n}+\dot{\varphi}_{n} \approx \Omega_{n}-\frac{\omega_{n}}{2\varepsilon_{o}}\,\mathcal{Re}\left\{\mathcal{P}_{n}\right\}/\mathcal{E}_{n}$$

Thus

$$\omega_n+\dot{\varphi}_n \qquad \Omega_n+\frac{1}{2}\,\omega_n\,\frac{{}^{2}}{\hbar\varepsilon_o}\,\frac{\left(\omega_{ab}-\omega_n\right)}{\gamma_{ab}^{2}}\,\mathcal{L}\left(\omega_{ab}-\omega_n;\gamma_{ab}\right)\,\bar{N}\left\{1-\frac{3}{4}\,I_n\left(\frac{\gamma_a+\gamma_b}{\gamma_{ab}}\right)\mathcal{L}\left(\omega_{ab}-\omega_n;\gamma_{ab}\right)\right\} \qquad [\text{ VI-27 }]$$

$$\Omega_n+\sigma_n-\beta_n\,I_n$$

where the linear mode ***pulling effect*** is measured by

$$\sigma_n=\frac{1}{2}\,\bar{N}\,\frac{\omega_n \quad {}^{2}}{\hbar\varepsilon_o\,\gamma_{ab}}\,\frac{\left(\omega_{ab}-\omega_n\right)}{\gamma_{ab}}\,\mathcal{L}\left(\omega_{ab}-\omega_n;\gamma_{ab}\right) \qquad [\text{ VI-28a }]$$

and nonlinear ***pushing effect*** by

$$\beta_n=\frac{3}{4}\,\bar{N}\,\frac{\omega_n \quad {}^{2}}{\hbar\varepsilon_o\,\gamma_{ab}}\,\frac{\left(\omega_{ab}-\omega_n\right)}{\gamma_{ab}}\left(\frac{\gamma_a+\gamma_b}{\gamma_{ab}}\right)\left[\mathcal{L}\left(\omega_{ab}-\omega_n;\gamma_{ab}\right)\right]^{2} \qquad [\text{ VI-28b }]$$

From Equation [VI-25a], we find the **"at-resonance"threshold condition to be:**

$$\left(\frac{\omega_n}{2Q_n}\right)=\frac{1}{2}\,\omega_n\,\frac{{}^{2}}{\hbar\varepsilon_o}\left(\frac{1}{\gamma_{ab}}\right)\mathcal{L}\left(0;\gamma_{ab}\right)\bar{N}_{Th} \qquad \frac{1}{Q_n}=\frac{{}^{2}\;\bar{N}_{Th}}{\hbar\varepsilon_o\,\gamma_{ab}} \qquad [\text{ VI-29 }]$$

where $\bar{N}_{Th}$ is the required population inversion at threshold.

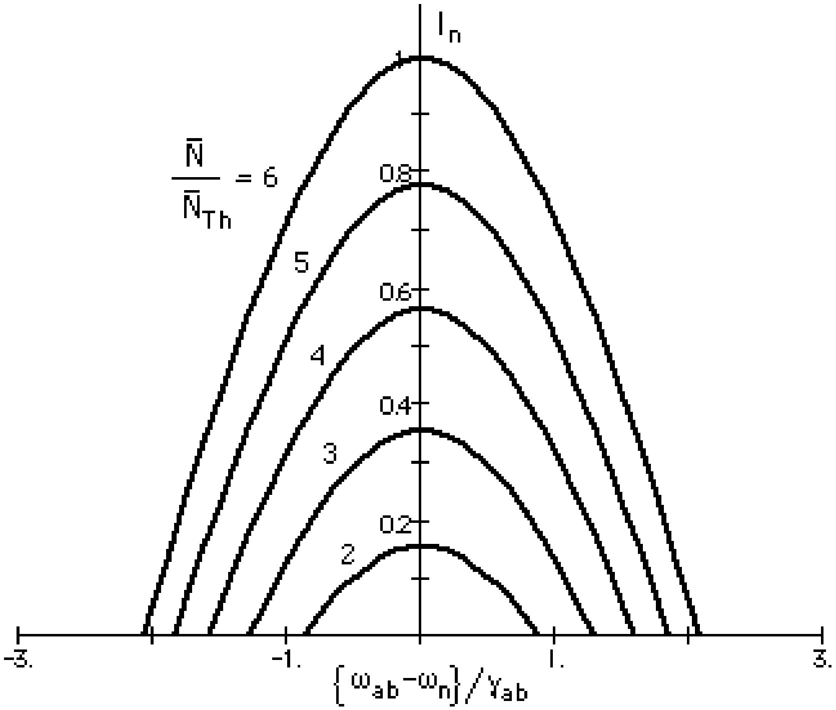
For the **general steady state condition**, we have

$$I_n=\frac{1}{2}\,\frac{{}^{2}\,\left|\mathcal{E}_n\right|^{2}}{\hbar^{2}}\,\frac{1}{\gamma_a\,\gamma_b}\approx\alpha_n/\beta_n \qquad [\text{ VI-30a }]$$

or

$$I_{\mathrm{n}} = \frac{4}{3}\left(\frac{\gamma_{ab}}{\gamma_a+\gamma_b}\right)\frac{\mathcal{L}\left(\omega_{ab}-\omega_{\mathrm{n}};\gamma_{ab}\right)-\bar{N}_{\mathrm{Th}}/\bar{N}}{\mathcal{L}^2\left(\omega_{ab}-\omega_{\mathrm{n}};\gamma_{ab}\right)} \qquad [\text{ VI-30b }]$$

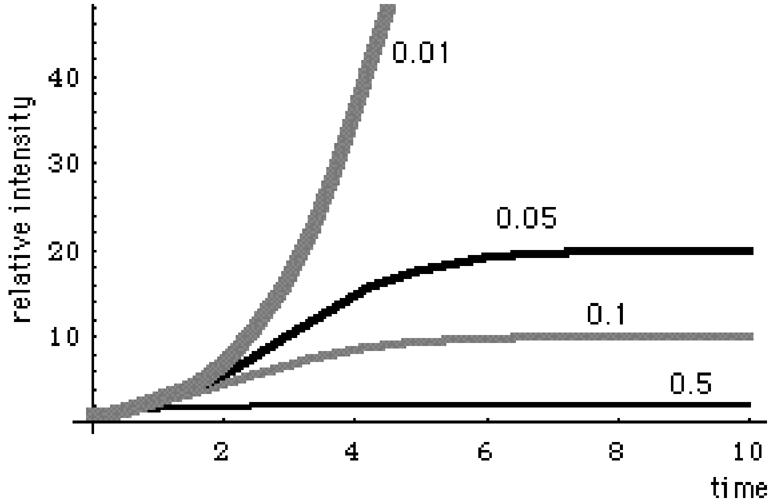
SINGLE-MODE, STANDING-WAVE LASER INTENSITY



The **time dependent intensity build-up** follows from integrating Equation [VI-25b] to obtain

$$I_{\mathrm{n}}(t) = \frac{\alpha_{\mathrm{n}}\, I_{\mathrm{n}}(0)\exp\left(\alpha_{\mathrm{n}}\, t\right)}{\alpha_{\mathrm{n}}+\beta_{\mathrm{n}}\, I_{\mathrm{n}}(0)\left[\exp\left(\alpha_{\mathrm{n}}\, t\right)-1\right]} \qquad [\text{ VI-29 }]$$

TRANSIENT INTENSITY BUILDUP



MULTIMODE OPERATION:

When we consider the possibility that two or more laser modes may be simultaneously excited or oscillating, things get quite a bit more complicated and we must refine our analysis. In particular, we must treat the time dependence of the population difference $\bar{\rho}_{aa} - \bar{\rho}_{bb}$ with greater care since variations in the population difference tend to modulate and couple the possible modal excitations.

Reflection on the form of Equations [VI-13a] and [VI-13b] in the absence of modal excitation, suggests that we take

$$N(z,t) \equiv \left[\bar{\rho}_{aa}^{(0)}(z,t) - \bar{\rho}_{bb}^{(0)}(z,t)\right] = \lambda_a \gamma_a^{-1} - \lambda_b \gamma_b^{-1} \qquad \text{[VI-31]}$$

as the zeroth-order approximation to the time dependence of the population difference. With finite excitation of one or more modes we may write in the rotating-wave approximation

$$\mathcal{V}_{ab}(z,t) = -\frac{1}{2} \sum_{n} \mathcal{E}_{n}(t) \exp\left[-i\left(\omega_{n}\, t + \varphi_{n}(t)\right)\right] U_{n}(z) \qquad [\text{ VI-32 }]$$

and use Equation [VI-14] to obtain the following first-order approximation to the time dependence of the off-diagonal component of the population matrix:

$$\begin{aligned} \rho^{(1)}_{ab} &= i\,\hbar^{-1} \int_{-\infty}^{t} d\,t' \exp\left[-\left(i\,\omega_{ab} + \gamma_{ab}\right)(t-t')\right] \mathcal{V}_{ab}(z,t')\left[\rho^{(0)}_{ab}(z,t) - \rho^{(0)}_{ab}(z,t)\right] \\ &= -\frac{1}{2} i \quad \hbar^{-1}\, N(z,t) \sum_{n} \mathcal{E}_{n}(t) \exp\left[-i\left(\omega_{n}\, t + \varphi_{n}(t)\right)\right] U_{n}(z) \int_{-\infty}^{t} d\,t' \exp\left\{-\left[i\left(\omega_{ab} - \omega_{n}\right) + \gamma_{ab}\right](t-t')\right\} \qquad [\text{ VI-33 }] \\ &= -\frac{1}{2} i \quad \hbar^{-1}\, N(z,t) \sum_{n} \mathcal{E}_{n}(t) \exp\left[-i\left(\omega_{n}\, t + \varphi_{n}(t)\right)\right] U_{n}(z)\, \mathcal{D}\left(\omega_{ab} - \omega_{n}; \gamma_{ab}\right) \end{aligned}$$

In carrying out this integration we have, again, assumed that changes in $\mathcal{E}_{n}$, and φ_{φ_n}, are negligible in a time γ^{-1}_{ab}. Using this result and Equation [VI-13a] we obtain the time derivative of the second-order approximation to the time dependence of the population of the upper level -- *viz*.

$$\begin{aligned} \dot{\rho}_{aa} &\quad \dot{\rho}^{(0)}_{aa} + \dot{\rho}^{(2)}_{aa} \\ &= \lambda_{a} - \gamma_{a}\, \rho_{aa} - \left[\left| \frac{}{2\hbar} \right|^{2} N(z,t) \sum_{n} \sum_{m} \mathcal{E}_{n} \mathcal{E}_{m} \exp\left\{ i\left[\left(\omega_{n} - \omega_{m}\right)t + \varphi_{n} - \varphi_{m}\right]\right\} U_{m}(z)\, U_{n}(z)\, \mathcal{D}\left(\omega_{ab} - \omega_{m}; \gamma_{ab}\right) + c.c. \right] \qquad [\text{ VI-34 }] \end{aligned}$$

The key point here is that the population ρ_{aa} has ***pulsations* at the intermode beat frequencies!** Integrating we find

$$\bar{\rho}_{aa}^{(2)} = -\left|\frac{\;}{2\hbar}\right|^2 N(z,t) \sum_{n}\sum_{m} \mathcal{E}_n \mathcal{E}_m \exp\left\{ i\left[(\omega_n - \omega_m)\, t + \varphi_n - \varphi_m\right]\right\} U_m(z)\, U_n(z)\, \mathcal{D}(\omega_n - \omega_m; \gamma_a)\, \mathcal{D}(\omega_{ab} - \omega_m; \gamma_{ab}) + c.c. \quad [\text{ VI-35 }]$$

and since $\bar{\rho}_{bb}^{(2)} = -\bar{\rho}_{aa}^{(2)}$ with $\gamma_a \quad \gamma_b$ we obtain the second-order approximation to the population difference as

$$\begin{aligned} \bar{\rho}_{aa}^{(2)} - \bar{\rho}_{bb}^{(2)} = -\left|\frac{\;}{2\hbar}\right|^2 N(z,t) \sum_{n}\sum_{m} \mathcal{E}_n \mathcal{E}_m \exp\left\{ i\left[(\omega_n - \omega_m)\, t + \varphi_n - \varphi_m\right]\right\} U_m(z)\, U_n(z) \\ \times\left[\mathcal{D}(\omega_n - \omega_m; \gamma_a) + \mathcal{D}(\omega_n - \omega_m; \gamma_b)\right]\left[\mathcal{D}(\omega_{ab} - \omega_m; \gamma_{ab}) + \mathcal{D}(\omega_n - \omega_{ab}; \gamma_{ab})\right] \end{aligned} \quad [\text{ VI-36 }]$$

Substituting this expression into the formal integral of Equation [VI-14], we find, directly, the third-order approximation to the off-diagonal component of the population matrix

$$\begin{aligned} \bar{\rho}_{ab}^{(3)} = \frac{1}{8}\, i\left|\frac{\;}{2\hbar}\right|^2 N(z,t) \sum_{l}\sum_{m}\sum_{n} \mathcal{E}_l \mathcal{E}_m \mathcal{E}_n\, U_l(z)\, U_m(z)\, U_n(z) \\ \times \exp\left\{-i\left[(\omega_l - \omega_m + \omega_n)\, t + \varphi_l - \varphi_m + \varphi_n\right]\right\} \mathcal{D}(\omega_{ab} - \omega_l + \omega_m - \omega_n; \gamma_{ab}) \\ \times\left[\mathcal{D}(\omega_m - \omega_n; \gamma_a) + \mathcal{D}(\omega_m - \omega_n; \gamma_b)\right]\left[\mathcal{D}(\omega_{ab} - \omega_n; \gamma_{ab}) + \mathcal{D}(\omega_m - \omega_{ab}; \gamma_{ab})\right] \end{aligned} \quad [\text{ VI-37 }]$$

Using Equation [VI-10b] and invoking Equations [VI-8] the general self-consistency conditions may be written [28]

---

[28] It is important to note, that use of Equation [VI-10b] requires evaluation of the following integral :

$$N_{klmn} = \int_0^L dz\, N(z)\, U_k(z)\, U_l(z)\, U_m(z)\, U_n(z) \Big/ \int_0^L dz\, |U_k(z)|^2$$

$$\dot{\mathcal{E}}_{\mathrm{k}} = a_{\mathrm{k}}\, \mathcal{E}_{\mathrm{k}} - \sum_{\mathrm{l}}\sum_{\mathrm{m}}\sum_{\mathrm{n}} \mathcal{E}_{\mathrm{l}}\mathcal{E}_{\mathrm{m}}\mathcal{E}_{\mathrm{n}}\ \mathrm{Im}\left\{\vartheta_{\mathrm{klmn}}\ \exp\left[\,i\,\Psi_{\mathrm{klmn}}\right]\right\} \qquad [\text{ VI-38 }]$$

$$\omega_{\mathrm{k}} + \dot{\varphi}_{\mathrm{k}} = \Omega_{\mathrm{k}} + \sigma_{\mathrm{k}} - \sum_{\mathrm{l}}\sum_{\mathrm{m}}\sum_{\mathrm{n}} \mathcal{E}_{\mathrm{k}}^{-1}\mathcal{E}_{\mathrm{l}}\mathcal{E}_{\mathrm{m}}\mathcal{E}_{\mathrm{n}}\ \mathrm{Re}\left\{\vartheta_{\mathrm{klmn}}\ \exp\left[\,i\,\Psi_{\mathrm{klmn}}\right]\right\} \qquad [\text{ VI-39 }]$$

with the coefficients summarized in the following table for a **standing wave** configuration:

Coefficients	**Significance**
$a_{\mathrm{k}} = \mathcal{L}\left(\omega_{ab} - \omega_{\mathrm{k}};\, \gamma_{ab}\right) F_1 - \tfrac{1}{2}\,\omega_{\mathrm{k}}/Q_{\mathrm{k}}$	**Linear net gain**
$\sigma_{\mathrm{k}} = \left[\left(\omega_{ab} - \omega_{\mathrm{k}}\right)/\gamma_{ab}\right] \mathcal{L}\left(\omega_{ab} - \omega_{\mathrm{k}};\, \gamma_{ab}\right) F_1$	**Linear mode pulling**
$\beta_{\mathrm{k}} = \mathcal{L}^2\left(\omega_{ab} - \omega_{\mathrm{k}};\, \gamma_{ab}\right) F_3$	**Self saturation**
$\vartheta_{\mathrm{klmn}} = i\,\frac{\gamma_{ab}}{4}\,\left\lvert\quad /2\hbar\right\rvert^2 F_1\left\{1 + \left[N_{2(\mathrm{m-n})} + N_{2(\mathrm{m-l})}\right]/\overline{N}\right\} \mathcal{D}\left(\omega_{ab} - \omega_{\mathrm{l}} + \omega_{\mathrm{m}} - \omega_{\mathrm{n}};\, \gamma_{ab}\right)$ $\times\left[\mathcal{D}\left(\omega_{ab} - \omega_{\mathrm{n}};\, \gamma_{ab}\right) + \mathcal{D}\left(\omega_{\mathrm{m}} - \omega_{ab};\, \gamma_{ab}\right)\right]\left[\mathcal{D}\left(\omega_{\mathrm{m}} - \omega_{\mathrm{n}};\, \gamma_{a}\right) + \mathcal{D}\left(\omega_{\mathrm{m}} - \omega_{\mathrm{n}};\, \gamma_{b}\right)\right]$	**General saturation term**

Thus, the coefficients in Equations [VI-38] and [VI-39] are sensitive to the particulars of the laser mode configuration. Since the modal excitations cannot change rapidly , the only integrals of importance are those for which $\omega_k - \omega_l + \omega_m - \omega_n \approx 0$

$$
\begin{aligned}
\vartheta_{kkmm} + \vartheta_{kmmk} = {} & i\,\frac{1}{2}\left[\frac{\gamma_a+\gamma_b}{\gamma_a\gamma_b}\right] \quad /2\hbar|^2\, F_1\left\{2+N_{2(m-k)}/\overline{N}\right\}\mathcal{D}\left(\omega_{ab}-\omega_k;\gamma_{ab}\right) \\
& \times\Big\{\mathcal{L}\left(\omega_{ab}-\omega_k;\gamma_{ab}\right)\mathcal{L}\left(\omega_{ab}-\omega_m;\gamma_{ab}\right) \\
& \qquad + \frac{\gamma_{ab}}{2}\left[\frac{\gamma_a\gamma_b}{\gamma_a+\gamma_b}\right]\left[\mathcal{D}\left(\omega_{ab}-\omega_k;\gamma_{ab}\right)+\mathcal{D}\left(\omega_m-\omega_{ab};\gamma_{ab}\right)\right] \\
& \qquad\qquad \times\left[\mathcal{D}\left(\omega_m-\omega_k;\gamma_a\right)+\mathcal{D}\left(\omega_m-\omega_k;\gamma_b\right)\right]\Big\}
\end{aligned}
$$

Stationary coefficient

$$
\begin{aligned}
\theta_{km} &= \left[\left(2\hbar^2\,\gamma_a\gamma_b\right)/|\quad|^2\right]\,\mathrm{Im}\left(\vartheta_{kkmm}+\vartheta_{kmmk}\right) \\
&= \frac{1}{3}F_3\left\{2+N_{2(m-k)}/\overline{N}\right\}\Big[\mathcal{L}\left(\omega_{ab}-\omega_k;\gamma_{ab}\right)\mathcal{L}\left(\omega_{ab}-\omega_m;\gamma_{ab}\right) \\
&\qquad + \frac{1}{2}\left[\gamma_{ab}\right]^2\left[\frac{\gamma_a\gamma_b}{\gamma_a+\gamma_b}\right]\mathrm{Re}\Big\{\mathcal{D}\left(\omega_{ab}-\omega_k;\gamma_{ab}\right)\left[\mathcal{D}\left(\omega_{ab}-\omega_k;\gamma_{ab}\right)+\mathcal{D}\left(\omega_m-\omega_{ab};\gamma_{ab}\right)\right] \\
&\qquad\qquad \times\left[\mathcal{D}\left(\omega_m-\omega_k;\gamma_a\right)+\mathcal{D}\left(\omega_m-\omega_k;\gamma_b\right)\right]\Big\}\Big]
\end{aligned}
$$

Cross-saturation

$$
\Psi_{klmn} = \left(\omega_k-\omega_l+\omega_m-\omega_n\right)\,t+\varphi_k-\varphi_l+\varphi_m-\varphi_n
$$

Relative phase angle

$$
\rho_m = \left(\omega_{ab}-\omega_m\right)/\gamma_{ab}\,\mathcal{L}^2\left(\omega_{ab}-\omega_m;\gamma_{ab}\right)F_3
$$

Self mode pushing

$$
\begin{aligned}
\tau_{km} &= \left[\left(2\hbar^2\,\gamma_a\gamma_b\right)/|\quad|^2\right]\,\mathrm{Re}\left(\vartheta_{kkmm}+\vartheta_{kmmk}\right) \\
&= \frac{1}{3}F_3\left\{2+N_{2(m-k)}/\overline{N}\right\}\Big[\left(\omega_{ab}-\omega_k\right)/\gamma_{ab}\;\mathcal{L}\left(\omega_{ab}-\omega_k;\gamma_{ab}\right)\mathcal{L}\left(\omega_{ab}-\omega_m;\gamma_{ab}\right) \\
&\qquad - \frac{1}{2}\left[\gamma_{ab}\right]^2\left[\frac{\gamma_a\gamma_b}{\gamma_a+\gamma_b}\right]\mathrm{Im}\Big\{\mathcal{D}\left(\omega_{ab}-\omega_k;\gamma_{ab}\right)\left[\mathcal{D}\left(\omega_{ab}-\omega_k;\gamma_{ab}\right)+\mathcal{D}\left(\omega_m-\omega_{ab};\gamma_{ab}\right)\right]
\end{aligned}
$$

Cross pushing

$F_1 = \omega_n \ ^2\, \overline{N} \left[2\hbar\varepsilon_o\, \gamma_{ab}\right]^{-1}$ $\quad$ $F_3 = \left(3/2\right) \dfrac{1}{\gamma_{ab}} \left[\dfrac{\gamma_a\gamma_b}{\gamma_a+\gamma_b}\right] F_1$	**First-order factor** and **Third-order factor**
$N_{2m} = \dfrac{1}{L}\displaystyle\int dz\ N\left(z\right) \cos\left[\left(\dfrac{2\pi m}{L}\right) z\right]$	**Spatial factor**
$\mathcal{D}\left(u-v;w\right) \equiv \left[i\left(u-v\right)+w\right]^{-1}$	**Lorentzian denominator**
$\mathcal{L}\left(u-v;w\right) \equiv w^2\left[\left(u-v\right)^2+w^2\right]^{-1}$	**Dimensionless Lorentzian**

Two-Mode Operation: For two modes the amplitude determining equations -- *i.e.* Equation [VI-38] - reduce to

$$\begin{aligned}\dot{\mathcal{E}}_1 &= \mathcal{E}_1\left[a_1-\beta_1 I_1-\theta_{12}\ I_2\right]\\ \dot{\mathcal{E}}_2 &= \mathcal{E}_2\left[a_2-\beta_2 I_2-\theta_{21}\ I_1\right]\end{aligned} \qquad \text{[VI-40]}$$

and the frequency determining equations -- *i.e.* Equation [VI-39] -- reduce to

$$\begin{aligned}\omega_1+\dot{\varphi}_1 &= \Omega_1+\sigma_1-\rho_1 I_1-\tau_{12}\ I_2\\ \omega_2+\dot{\varphi}_2 &= \Omega_2+\sigma_2-\rho_2\ I_2-\tau_{21}\ I_1 w\end{aligned} \qquad \text{[VI-41]}$$

Multiplying the amplitude equations by $\mathcal{E}_s| \ \ |^2/\left(\hbar^2\gamma_a\gamma_b\right)$ we obtain the equations of motion for the dimensionless intensities

$$\begin{aligned}\dot{I}_1 &= 2\ I_1\left[a_1-\beta_1\ I_1-\theta_{12}\ I_2\right]\\ \dot{I}_2 &= 2\ I_2\left[a_2-\beta_2\ I_2-\theta_{21}\ I_1\right]\end{aligned} \qquad \text{[VI-42]}$$

STABILITY OF POSSIBLE STEADY STATE SOLUTIONS:

Stability criterion for stationary solutions: If $I_1 = I_1^{(s)} + \varepsilon$ and $I_2 = I_2^{(s)} + \varepsilon_2$, $\varepsilon_1 \to 0$ and $\varepsilon_2 \to 0$ as $t \to \infty$.

Solution 1: $I_1^{(s)} = 0$ and $I_2^{(s)} = a_2/\beta_2$ so that

$$\begin{aligned} \varepsilon_1 &= 2[\varepsilon_1]\left[a_1 - \beta_1 \varepsilon_1 - \theta_{12}\left(a_2/\beta_2 + \varepsilon_2\right)\right] \\ \varepsilon_2 &= 2\left[a_2/\beta_2 + \varepsilon_2\right]\left[a_2 - \beta_2\left(a_2/\beta_2 + \varepsilon_2\right) - \theta_{21}\varepsilon_1\right] \end{aligned} \qquad \text{[VI-43a]}$$

or

$$\begin{aligned} \dot{\varepsilon}_1 &= 2\,\varepsilon_1\left[a_1 - \theta_{12}\, a_2/\beta_2\right] + O\left(\varepsilon^2\right) \\ \dot{\varepsilon}_2 &= -2\left(a_2/\beta_2\right)\left[\beta_2\,\varepsilon_2 + \theta_{21}\,\varepsilon_1\right] + O\left(\varepsilon^2\right) \end{aligned} \qquad \text{[VI-43b]}$$

Stability requires that $a_1' \equiv a_1 - \theta_{12}\, a_2/\beta_2$ remain negative. If $a_1' < 0$ but $a_1 > 0$ one says that I_2 inhibits the oscillation of I_1 (**mode inhibition**). If a_1 becomes large to overcome the inhibiting effect of I_2, the solution becomes unstable and I_1 builds up.

Solution 2: $I_1^{(s)} \neq 0$ and $I_2^{(s)} \neq 0$ so that

$$\begin{aligned} a_1 &= \beta_1\, I_1^{(s)} + \theta_{12}\, I_2^{(s)} \\ a_2 &= \beta_2\, I_2^{(s)} + \theta_{21}\, I_1^{(s)} \end{aligned} \qquad \text{[VI-44]}$$

which has the solution

$$\begin{aligned} I_1^{(s)} &= \frac{\left(a_1/\beta_1\right) - \left(\theta_{21}/\beta_1\right)\left(a_2/\beta_2\right)}{1-C} = \frac{a_1'/\beta_1}{1-C} \\ I_2^{(s)} &= \frac{\left(a_2/\beta_2\right) - \left(\theta_{12}/\beta_2\right)\left(a_1/\beta_1\right)}{1-C} = \frac{a_2'/\beta_2}{1-C} \end{aligned} \qquad \text{[VI-45]}$$

where the ***coupling constant*** $C = \frac{\theta_{12}\,\theta_{21}}{\beta_1\,\beta_2}$. Substituting this solution into Equation [VI-42] we see that

$$\begin{aligned} \dot{\varepsilon}_1 &= -2\, I_1^{(s)} \left[\beta_1\, \varepsilon_1 + \theta_{12}\, \varepsilon_2\right] + \mathrm{O}\left(\varepsilon^2\right) \\ \dot{\varepsilon}_2 &= -2\, I_2^{(s)} \left[\beta_2\, \varepsilon_2 + \theta_{21}\, \varepsilon_1\right] + \mathrm{O}\left(\varepsilon^2\right) \end{aligned} \qquad \text{[VI-46a]}$$

In matrix form

$$\frac{d}{dt}\begin{bmatrix}\varepsilon_1\\ \varepsilon_2\end{bmatrix} = \frac{-2}{1-C}\begin{bmatrix} a_1' & a_1'\left(\theta_{12}/\beta_1\right) \\ a_2'\left(\theta_{21}/\beta_2\right) & a_2' \end{bmatrix}\begin{bmatrix}\varepsilon_1\\ \varepsilon_2\end{bmatrix} = \mathcal{M}\begin{bmatrix}\varepsilon_1\\ \varepsilon_2\end{bmatrix} \qquad \text{[VI-46b]}$$

The solutions will be stable if the eigenvalues of this equation are both negative.

$$\mathcal{M}\begin{bmatrix}\varepsilon_1\\ \varepsilon_2\end{bmatrix} = \lambda\begin{bmatrix}\varepsilon_1\\ \varepsilon_2\end{bmatrix} \qquad \mathrm{Det}\left(\mathcal{M} - \lambda\, I\right) = 0 \qquad \text{[VI-47]}$$

$$\lambda_{\left(\substack{1\\2}\right)} = -\frac{a_1' + a_2'}{1-C} \pm \sqrt{\left(\frac{a_1' + a_2'}{1-C}\right)^2 - 4\left(\frac{a_1'\,a_2'}{1-C}\right)} \qquad \text{[VI-48]}$$

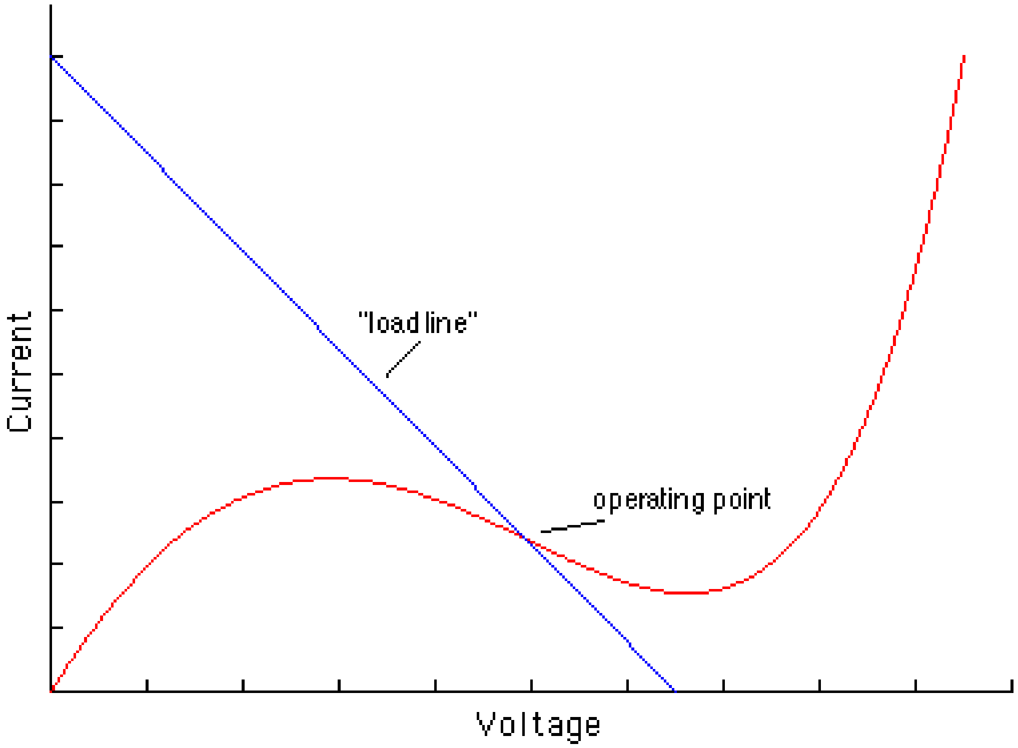
Case 1: **Weak coupling** where $C < 1$ and $a_1' > 0$ and $a_2' > 0$: **Stable** since both eigenvalues negative

Case 2: **Strong coupling** $C > 1$ and $a_1' < 0$ and $a_2' < 0$: **Unstable** since one eigenvalue is positive

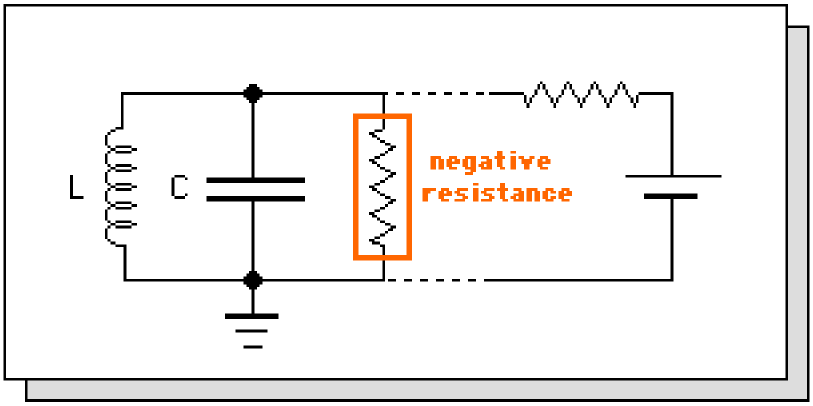
The van der Pol Negative Resistance Oscillator

Van der Pol's analysis[1] of "negative resistance" (*e.g.*, tunnel diode) oscillators prides a valuable framework for treating relative simplicity important features of oscillatory systems.

The characteristic curve of a "negative resistance" device



Consider the following negative resistance oscillatory circuit:



By simple circuit analysis, it is a straightforword proposition to find the following simple circuit equation which is the fundamental van der Pol oscillator equation:

---

[1] B. van der Pol, *Radio Rev.* **1**, 704-754, 1920 and B. van der Pol, *Phil. Mag.* **3**, 65, 1927

$$\frac{d^2}{d\,t^2}\,v\,(t)-\frac{d}{d\,t}\Big[\alpha\;v(t)-\bar{\beta}\;v^3(t)\Big]+\omega_0^2\;v\,(t)=0 \qquad \text{[VdP-1]}$$

where $\omega_0^2=\left(\mathrm{LC}\right)^{-1}$.

If α is small, it is reasonable to take

$$v\,(t)=\frac{1}{2}\,V(t)\,\exp\!\left(-i\,\omega_0\,t\right)+c.c. \qquad \text{[VdP-2]}$$

Then Equation [VdP-1] becomes without approximation

$$\begin{aligned}
&\left\{\frac{1}{2}\Big[-\omega_0^2\,V(t)-i\,2\,\omega_0\,\dot{V}(t)+\ddot{V}(t)\Big]\,\exp\!\left(-i\,\omega_0\,t\right)+c.c.\right\}\\
&\quad-\Big[\alpha-3\,\bar{\beta}\,v^2(t)\Big]\left\{\frac{1}{2}\Big[-i\,\omega_0\,V(t)+\dot{V}(t)\Big]\,\exp\!\left(-i\,\omega_0\,t\right)+c.c.\right\}\\
&\quad+\omega_0^2\left\{\frac{1}{2}\,V(t)\,\exp\!\left(-i\,\omega_0\,t\right)+c.c.\right\}=0
\end{aligned} \qquad \text{[VdP-3]}$$

If we ignore harmonic generation, Equation [VdP-3] may be approximated as

$$\begin{aligned}
&\left\{\frac{1}{2}\Big[-i\,2\,\omega_0\,\dot{V}(t)+\ddot{V}(t)\Big]\,\exp\!\left(-i\,\omega_0\,t\right)+c.c.\right\}\\
&\quad-\alpha\left\{\frac{1}{2}\Big[-i\,\omega_0\,V(t)+\dot{V}(t)\Big]\,\exp\!\left(-i\,\omega_0\,t\right)+c.c.\right\}\\
&\quad+\frac{3}{8}\,\bar{\beta}\,\Bigg[\Bigg\{-i\,\omega_0\,|\,V(t)\,|^2\;V(t)+\frac{d}{d\,t}\Big[|\,V(t)\,|^2\;V(t)\Big]\Bigg\}\exp\!\left(-i\,\omega_0\,t\right)+c.c.\Bigg]=0
\end{aligned} \qquad \text{[VdP-4]}$$

If we make the **slow time variation** assumption, this equation reduces to

$$\dot{V}(t)=\frac{1}{2}\left[\alpha-\frac{3}{4}\,\bar{\beta}\,|\,V(t)\,|^2\right]V(t)=\frac{1}{2}\Big[\alpha-\beta\,|\,V(t)\,|^2\Big]\;V(t) \qquad \text{[VdP-5]}$$

where $\beta=3\,\bar{\beta}/4$. This essential Equation [VI-25b] in the lecture set entitled *The Interaction of Radiation and Matter: Semiclassical Theory*. We saw there that the general steady state solution is given by

$$|\,V(t)\,|^2=\frac{\alpha}{\beta} \qquad \text{[VdP-6]}$$

To study **frequency locking** we suppose that a driving source (to be precise a current source in parallel with the negative resistance) and then the van der Pol equation becomes

$$\frac{d^2}{d\,t^2}\,v\,(t) - \frac{d}{d\,t}\left[\alpha\;v(t) - \bar{\beta}\;v^3(t)\right] + \omega_0^2\;v\,(t) = \omega^2\,V_0\,\sin\omega\,t$$

$$= \frac{\omega^2\,V_0}{2}\left[i\,\exp\left(-i\,\omega\,t\right) + c.c.\right] \qquad [\text{ VdP-7 }]$$

In this case, it is reasonable to take

$$v\,(t) = \frac{1}{2}\,V(t)\,\exp\left(-i\,\omega\,t\right) + c.c. \qquad [\text{ VdP-8 }]$$

If we again ignore harmonic geneation, Equation [VdP-7] becomes

$$\begin{aligned}
&\left\{\frac{1}{2}\left[-\omega^2\,V(t) - i\;2\,\omega\,\dot{V}(t) + \ddot{V}(t)\right]\,\exp\left(-i\,\omega t\right) + c.c.\right\}\\
&\quad -\left[\alpha - 3\,\bar{\beta}\,v^2\!\left(t\right)\right]\left\{\frac{1}{2}\left[-i\,\omega V(t) + \dot{V}(t)\right]\,\exp\left(-i\,\omega t\right) + c.c.\right\}\\
&\quad +\omega_0^2\left\{\frac{1}{2}\,V(t)\,\exp\left(-i\,\omega t\right) + c.c.\right\}\\
&= \frac{\omega^2\,V_0}{2}\left[i\,\exp\left(-i\,\omega\,t\right) + c.c.\right]
\end{aligned} \qquad [\text{ VdP-9 }]$$

Again under the **slow time variation** assumption, this equation reduces to

$$\frac{1}{2}\left(\omega_0^2 - \omega^2\right)V(t) - i\,\omega\dot{V}(t) + i\,\omega\,\frac{1}{2}\left[\alpha - \beta\,|\,V(t)\,|^2\right]V(t) = i\,\frac{\omega^2\,V_0}{2} \qquad [\text{ VdP-10 }]$$

If we take $V(t) = |\,V(t)\,|\,\exp\left(-i\,\Psi(t)\right)$, this equation separate into the following pair of equations:

$$|\,\dot{V}(t)\,| = \frac{1}{2}\left[\alpha - \beta\,|\,V(t)\,|^2\right]|\,V(t)\,| - \frac{\omega V_0}{2}\cos\Psi(t) \qquad [\text{ VdP-11a }]$$

$$\dot{\Psi}(t) = \frac{1}{2}\,\frac{\left(\omega_0^2 - \omega^2\right)}{\omega} + \frac{\omega}{2}\,\frac{V_0}{|\,V(t)\,|}\,\sin\Psi(t) = d + l\,\sin\Psi(t) \qquad [\text{ VdP-11b }]$$

where $d \equiv \frac{1}{2}\,\frac{\left(\omega_0^2 - \omega^2\right)}{\omega} = \left(\omega_0 - \omega\right)$ (the “detuning term”) and $l \equiv \frac{\omega}{2}\,\frac{V_0}{|\,V(t)\,|}$ (the “locking coefficient"). For small V_0 we can decouple the equations and take $|V(t)|^2 = \frac{\alpha}{\beta}$ from Equation [VdP-6] so that $l \equiv \frac{\omega}{2}\frac{V_0}{|V(t)|} = \frac{\omega}{2}\frac{\beta}{\alpha}V_0$. If $|d/l| >> 1$ the relative phase angle changes linearly in time at the rate $\dot{\Psi} \approx d$. As $|d/l|$ decreases toward unity, the "locking term" subtracts from the "detuning term" in one half of a cycle and adds in the other half. At $|d/l| \le 1$ there are two values of the phase angle that yield the "mode locking" condition $\dot{\Psi} = 0$ -- *viz*.

$$\Psi_{\mathrm{PL}} = \begin{cases} -\sin^{-1}(d/l) \\ \pi + \sin^{-1}(d/l) \end{cases} \qquad \text{[VdP-12]}$$

We can test the stability of these solutions by taking $\Psi(t) = \Psi_{\mathrm{PL}} + \varepsilon(t)$ and therefore Equation [VdP-11b] becomes

$$\dot{\varepsilon}(t) \approx l\cos(\Psi_{\mathrm{PL}})\,\varepsilon(t) \qquad \text{[VdP-13]}$$

and the solutions are stable if

$$l\cos(\Psi_{\mathrm{PL}}) < 0 \qquad \text{[VdP-14a]}$$

$$\sqrt{l^2 - d^2} < 0 \qquad \text{[VdP-14b]}$$

The van der Pol Negative Resistance Oscillator

VII. SEMICONDUCTOR PHOTONICS

A. PRELIMINARIES: SEMICONDUCTOR BACKGROUND[29]

THE CRYSTAL HAMILTONIAN

For an assembly of atoms the classical energy is the sum of the following:

- the kinetic energy of the nuclei;
- the potential energy of the nuclei in one another's electrostatic field;
- the kinetic energy of the electrons;
- the potential energy of the electrons in the field of the nuclei;
- the potential energy of the electrons in one another's electrostatic field;
- the magnetic energy associated with spin and orbital variables.

Dividing the electrons into core and valence electrons and leaving out magnetic effects leads to the following expression for the crystal Hamiltonian:

$$\mathcal{H} = \sum_{\alpha} \frac{\vec{\mathbf{p}}_{\alpha}^{\,2}}{2M_{\alpha}} + \sum_{\alpha\,\beta} U\left(\vec{\mathbf{R}}_{\alpha} - \vec{\mathbf{R}}_{\beta}\right) + \sum_{l} \frac{\vec{\mathbf{p}}_{l}^{\,2}}{2m_{l}} + \sum_{l,\,\alpha} V\left(\vec{\mathbf{r}}_{l} - \vec{\mathbf{R}}_{\alpha}\right) + \sum_{l,\,m} \frac{e^{2}/4\pi\varepsilon_{0}}{\left|\vec{\mathbf{r}}_{l} - \vec{\mathbf{r}}_{m}\right|} \qquad [\text{ VII-0 }]$$

where α and β label the ions, l and m label the electrons, $\vec{\mathbf{p}}$ is the momentum, M is an ionic mass, m is the mass of an electron, $U\left(\vec{\mathbf{R}}_{\alpha} - \vec{\mathbf{R}}_{\beta}\right)$ is the interionic potential, and $V\left(\vec{\mathbf{r}}_{l} - \vec{\mathbf{R}}_{\alpha}\right)$ is the valence-electron-ion potential.

The quantum mechanics of the assembly is treated to a good approximation by taking the total wavefunction of the system as the product

$$\Xi = \Psi\left(\vec{\mathbf{r}};\vec{\mathbf{R}}\right)\Phi\left(\vec{\mathbf{R}}\right) \qquad [\text{ VII-1 }]$$

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[29] This discussion draws heavily on B. K. Ridley, *Quantum Processes in Semiconductors* (3rd edition), Clarendon Press (1993).

where $\Phi\left(\vec{\mathbf{R}}\right) = \Phi\left(\vec{\mathbf{R}}_\alpha, \vec{\mathbf{R}}_\beta, \vec{\mathbf{R}}_\delta, \vec{\mathbf{R}}_\gamma, \ldots\right)$ is the wavefunction of all the ions and $\Psi\left(\vec{\mathbf{r}}; \vec{\mathbf{R}}\right) = \Psi\left(\vec{\mathbf{r}}_k, \vec{\mathbf{r}}_l, \vec{\mathbf{r}}_m, \vec{\mathbf{r}}_n, \ldots; \vec{\mathbf{R}}_\alpha, \vec{\mathbf{R}}_\beta, \vec{\mathbf{R}}_\delta, \vec{\mathbf{R}}_\gamma, \ldots\right)$ is the wavefunction of all the electrons at the instantaneous ionic positions. The Schrödinger equation is then written

$$\begin{aligned}\Psi\left(\vec{\mathbf{r}};\vec{\mathbf{R}}\right) \mathcal{H}_{lattice}\, \Phi\left(\vec{\mathbf{R}}\right) + \Phi\left(\vec{\mathbf{R}}\right) \mathcal{H}_{elec}\, \Psi\left(\vec{\mathbf{r}};\vec{\mathbf{R}}\right) \\ + \left[\mathcal{H}_{lattice}\, \Psi\left(\vec{\mathbf{r}};\vec{\mathbf{R}}\right) \Phi\left(\vec{\mathbf{R}}\right) - \Psi\left(\vec{\mathbf{r}};\vec{\mathbf{R}}\right) \mathcal{H}_{lattice}\, \Phi\left(\vec{\mathbf{R}}\right)\right] = E\, \Psi\left(\vec{\mathbf{r}};\vec{\mathbf{R}}\right) \Phi\left(\vec{\mathbf{R}}\right)\end{aligned} \qquad [\text{ VII-2 }]$$

where the total Hamiltonian is parsed into two independent components -- *viz.*

$$\mathcal{H}_{lattice} = \sum_\alpha \frac{\vec{\mathbf{p}}_\alpha^{\,2}}{2M_\alpha} + \sum_{\alpha,\beta} U\left(\vec{\mathbf{R}}_\alpha - \vec{\mathbf{R}}_\beta\right) . \qquad [\text{ VII-3a }]$$

$$\mathcal{H}_{elec} = \sum_l \frac{\vec{\mathbf{p}}_l^{\,2}}{2m_l} + \sum_{l,\alpha} V\left(\vec{\mathbf{r}}_l - \vec{\mathbf{R}}_\alpha\right) + \sum_{l,m} \frac{e^2/4\pi\varepsilon_0}{\left|\vec{\mathbf{r}}_l - \vec{\mathbf{r}}_m\right|} . \qquad [\text{ VII-3b }]$$

The essential assumption of the ***adiabatic approximation*** is that the bracketed term is negligible and that the global problem may be treated as two independent problems -- *viz.*

$$\mathcal{H}_{lattice}\, \Phi\left(\vec{\mathbf{R}}\right) = E_{lattice}\, \Phi\left(\vec{\mathbf{R}}\right) \qquad [\text{ VII-4a }]$$

$$\mathcal{H}_{elec}\, \Psi\left(\vec{\mathbf{r}};\vec{\mathbf{R}}\right) = E_{elec}\, \Psi\left(\vec{\mathbf{r}};\vec{\mathbf{R}}\right) \qquad [\text{ VII-4b }]$$

As a further refinement, the electron problem must be parsed once more as

$$\mathcal{H}_{elec} = \left\{\mathcal{H}_{elec}\right\}_{static} + \left\{\mathcal{H}_{elec}\right\}_{dynamic}$$

where $\{\mathcal{H}_{elec}\}_{static}$ defines the problem of the many electron system interacting with the static ionic lattice and $\{\mathcal{H}_{elec}\}_{dynamic}$ incorporates the effects of the electron-phonon interaction.

LATTICE VIBRATIONS

For a brief overview of lattice vibrations see the webpage entitled *Self Study Materials on Solid State Electronics* 2 at

http://deas.harvard.edu/courses/ap216/lectures/ls_2/ls2_u7/sse_tut_2/solid2.html

BLOCH ELECTRONS - Solutions of the Schrödinger equation for a single electron or quasi-particle moving in a periodic lattice

If the electron-electron is averaged out, deviations from the average may be treated perturbations -- *i.e.*, we make the replacement

$$\sum_{l,\,m} \frac{e^2/4\pi\varepsilon_0}{\left|\vec{\mathbf{r}}_l - \vec{\mathbf{r}}_m\right|} \qquad \left(\mathcal{H}_{elec}\right)_0 + \left(\mathcal{H}_{elec}\right)_1 \qquad \text{[VII-5]}$$

where $\left(\mathcal{H}_{elec}\right)_0$ leads to a constant replusive electronic energy component and $\left(\mathcal{H}_{elec}\right)_1$ is a small fluctuating electron-electron interaction. If we neglect these fluctuations, each electron interacts independently with a screened lattice potential. In this approximation, the electronic wavefunction can be expressed as

$$\Psi\left(\vec{\mathbf{r}};\vec{\mathbf{R}}_0\right) = \prod_{\mathrm{n}} \psi_{\mathrm{n}}\left(\vec{\mathbf{r}}_{\mathrm{n}};\vec{\mathbf{R}}_0\right) \qquad \text{[VII-6]}$$

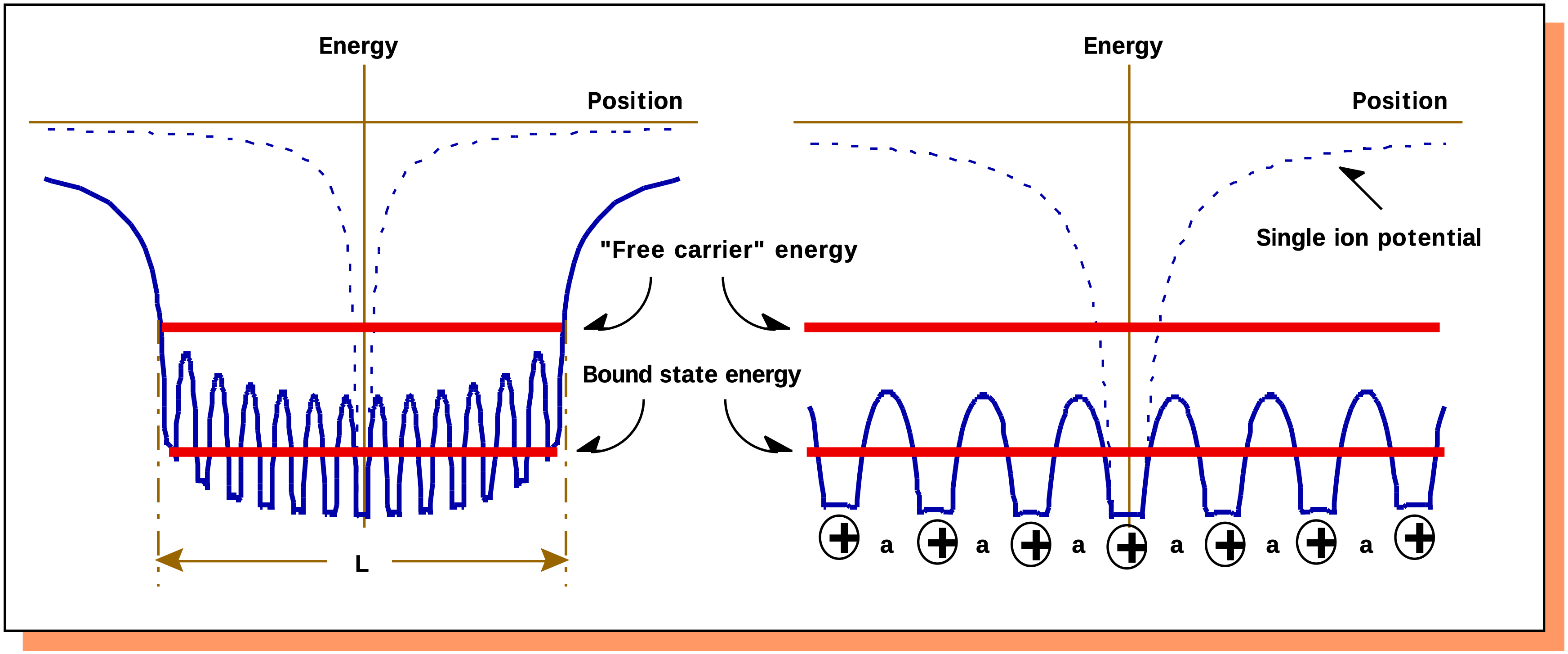
with the requirement that the occupation of one-electron states is in accordance with Pauli exclusion principle. Thus, each the wavefunction for each electrons satisfies a Schrödinger equation in the form

$$\left(\mathcal{H}^{\,n}_{elec}\right)_{static}\ \psi_n\left(\vec{\mathbf{r}}_n;\vec{\mathbf{R}}_0\right)=-\frac{\hbar^2}{2m}\nabla^2_n\ \psi_n\left(\vec{\mathbf{r}}_n;\vec{\mathbf{R}}_0\right)+V\left(\vec{\mathbf{r}}\right)\ \psi_n\left(\vec{\mathbf{r}}_n;\vec{\mathbf{R}}_0\right)=\mathcal{E}_n\ \psi_n\left(\vec{\mathbf{r}}_n;\vec{\mathbf{R}}_0\right)\qquad [\text{ VII-7a }]$$

or simplifying the notation

$$-\frac{\hbar^2}{2m}\nabla^2\ \psi\left(\vec{\mathbf{r}}\right)+V\left(\vec{\mathbf{r}}\right)\ \psi\left(\vec{\mathbf{r}}\right)=\mathcal{E}\ \psi\left(\vec{\mathbf{r}};\right)\qquad [\text{ VII-7b }]$$

SCHEMATIC PERIODIC LATTICE POTENTIAL



The lattice potential far from the surface of the crystal has the property that, for all lattice vectors $\vec{\mathbf{R}}_{[\ell]} = \ell_1\,\vec{\mathbf{a}}_1 + \ell_2\,\vec{\mathbf{a}}_2 + \ell_3\,\vec{\mathbf{a}}_3$

$$V\left(\vec{\mathbf{r}}\right) = V\left(\vec{\mathbf{r}} + \vec{\mathbf{R}}_{[\ell]}\right) \qquad \text{[VII-8]}$$

Bloch's (Floquet's) theorem: The function

$$\psi_{\vec{\mathbf{k}}}\left(\vec{\mathbf{r}}\right) = \left(\mathcal{V}\right)^{-1/2}\, u_{\vec{\mathbf{k}}}\left(\vec{\mathbf{r}}\right)\, \exp\left(i\,\vec{\mathbf{k}}\;\vec{\mathbf{r}}\right) \qquad \text{[VII-9]}$$

where $u_{\vec{\mathbf{k}}}\left(\vec{\mathbf{r}}\right)$ is a function with the same spatial periodicity as $V\left(\vec{\mathbf{r}}\right)$ that is an eigenfunction of Equation [VII-7].

Proof of theorem:

First rewrite Equation [VII-7b] as

$$\mathcal{H}\left(\vec{\mathbf{r}}\right)\,\psi\left(\vec{\mathbf{r}}\right) = \mathcal{E}\,\psi\left(\vec{\mathbf{r}}\right) \qquad \text{[VII-7b']}$$

Shifting the origin by a lattice vector, we again rewrite Equation [VII-7b] as

$$\mathcal{H}\left(\vec{\mathbf{r}} + \vec{\mathbf{R}}_{[\ell]}\right)\,\psi\left(\vec{\mathbf{r}} + \vec{\mathbf{R}}_{[\ell]}\right) = \mathcal{E}\,\psi\left(\vec{\mathbf{r}} + \vec{\mathbf{R}}_{[\ell]}\right) \qquad \text{[VII-7b'']}$$

In light of periodicity expressed in Equation [VII-8], it follows that

$$\mathcal{H}\left(\vec{\mathbf{r}}\right)\,\psi\left(\vec{\mathbf{r}} + \vec{\mathbf{R}}_{[\ell]}\right) = \mathcal{E}\,\psi\left(\vec{\mathbf{r}} + \vec{\mathbf{R}}_{[\ell]}\right) \qquad \text{[VII-10]}$$

Thus, any linear combination of the possible eigenfunctions $\psi\left(\vec{\mathbf{r}} + \vec{\mathbf{R}}_{[\ell]}\right)$ is a valid eigenstate of the energy $\mathcal{E}$! In particular, let us choose the combination

$$\begin{aligned}\psi_{\vec{\mathbf{k}}}(\vec{\mathbf{r}}) &= \sum_{[\ell]} \psi\left(\vec{\mathbf{r}}+\vec{\mathbf{R}}_{[\ell]}\right) \exp\left(-i\,\vec{\mathbf{k}}\;\;\vec{\mathbf{R}}_{[\ell]}\right) \\ &= \exp\left(\;i\,\vec{\mathbf{k}}\;\;\vec{\mathbf{r}}\right) \sum_{[\ell]} \psi\left(\vec{\mathbf{r}}+\vec{\mathbf{R}}_{[\ell]}\right) \exp\left[-i\,\vec{\mathbf{k}}\;\;\left(\vec{\mathbf{r}}+\vec{\mathbf{R}}_{[\ell]}\right)\right]\end{aligned} \qquad [\text{ VII-11 }]$$

where $\vec{\mathbf{k}}$ is, for the moment, taken to be an arbitrary **complex** vector. Since the summation includes all possible lattice vectors it must be a periodic function of $\vec{\mathbf{r}}$ with the period of the lattice and may be identified with $u_{\vec{\mathbf{k}}}(\vec{\mathbf{r}})$ of Equation [VII-9].[30] If we impose cyclical boundary conditions on $\psi_{\vec{\mathbf{k}}}(\vec{\mathbf{r}})$, we see that, of necessity, $\vec{\mathbf{k}}$ is a real vector! **QED**

In light of Bloch's theorem, Equation [VII-7b] may be rewrite in the form

$$\mathcal{H}_{\vec{\mathbf{k}}}(\vec{\mathbf{r}})\; u_{\vec{\mathbf{k}}}(\vec{\mathbf{r}}) = \left[-\frac{\hbar^2}{2m}\left(\;\; + i\,\vec{\mathbf{k}}\right)^2 + V(\vec{\mathbf{r}})\right] u_{\vec{\mathbf{k}}}(\vec{\mathbf{r}}) = \varepsilon_{\vec{\mathbf{k}}}\; u_{\vec{\mathbf{k}}}(\vec{\mathbf{r}})\;. \qquad [\text{ VII-12 }]$$

Since the reciprocal lattice vectors of crystal are defined[31] so that $\exp\left(\,i\,\vec{\mathbf{R}}_{[\ell]}\;\;\vec{\mathbf{G}}_{[m]}\right) = 1$, any function written in the form

$$\sum_{[m]} C_{[m]}\; \exp\left(\;i\,\vec{\mathbf{G}}_{[m]}\;\;\vec{\mathbf{r}}\right)$$

---

[30] We keep for the record $\quad u_{\vec{\mathbf{k}}}(\vec{\mathbf{r}}) \quad \sum_{[\ell]} \psi\left(\vec{\mathbf{r}}+\vec{\mathbf{R}}_{[\ell]}\right) \exp\left[-i\,\vec{\mathbf{k}}\;\;\left(\vec{\mathbf{r}}+\vec{\mathbf{R}}_{[\ell]}\right)\right]$

[31] The reciprocal lattice vectors: $\quad \vec{\mathbf{G}}_{[m]} = m_1\,\vec{\mathbf{b}}_1 + m_2\,\vec{\mathbf{b}}_2 + m_3\,\vec{\mathbf{b}}_3$

where

$$\vec{\mathbf{b}}_1 = 2\pi\frac{\vec{\mathbf{a}}_2\times\vec{\mathbf{a}}_3}{\vec{\mathbf{a}}_1\;\;\vec{\mathbf{a}}_2\times\vec{\mathbf{a}}_3}\,;\quad \vec{\mathbf{b}}_2 = 2\pi\frac{\vec{\mathbf{a}}_3\times\vec{\mathbf{a}}_1}{\vec{\mathbf{a}}_1\;\;\vec{\mathbf{a}}_2\times\vec{\mathbf{a}}_3}\,;\quad \vec{\mathbf{b}}_3 = 2\pi\frac{\vec{\mathbf{a}}_1\times\vec{\mathbf{a}}_2}{\vec{\mathbf{a}}_1\;\;\vec{\mathbf{a}}_2\times\vec{\mathbf{a}}_3}$$

has the spatial periodicity of the lattice. Conversely, we are allowed to expand $V(\vec{\mathbf{r}})$ and $u_{\vec{\mathbf{k}}}(\vec{\mathbf{r}})$ in Fourier series -- *viz.*

$$V(\vec{\mathbf{r}}) = \sum_{[m]} V_{[m]} \exp\left(i\, \vec{\mathbf{G}}_{[m]} \quad \vec{\mathbf{r}} \right) \qquad \text{[VII-13a]}$$

$$u_{\vec{\mathbf{k}}}(\vec{\mathbf{r}}) = \sum_{[m']} u_{[m']}(\vec{\mathbf{k}}) \exp\left(i\, \vec{\mathbf{G}}_{[m']} \quad \vec{\mathbf{r}} \right) \qquad \text{[VII-14a]}$$

where

$$V_{[m]} = \mathcal{V}^{-1} \iiint d\vec{\mathbf{r}}\, V(\vec{\mathbf{r}}) \exp\left(-i\, \vec{\mathbf{G}}_{[m']} \quad \vec{\mathbf{r}} \right) \qquad \text{[VII-13b]}$$

and

$$u_{[m]}(\vec{\mathbf{k}}) = \mathcal{V}^{-1} \iiint d\vec{\mathbf{r}}\, u_{\vec{\mathbf{k}}}(\vec{\mathbf{r}}) \exp\left(-i\, \vec{\mathbf{G}}_{[m]} \quad \vec{\mathbf{r}} \right) \qquad \text{[VII-14b]}$$

Substituting these expressions into Equation [VII-12] and equating various Fourier components, we obtain the following infinite set of algebraic equations:

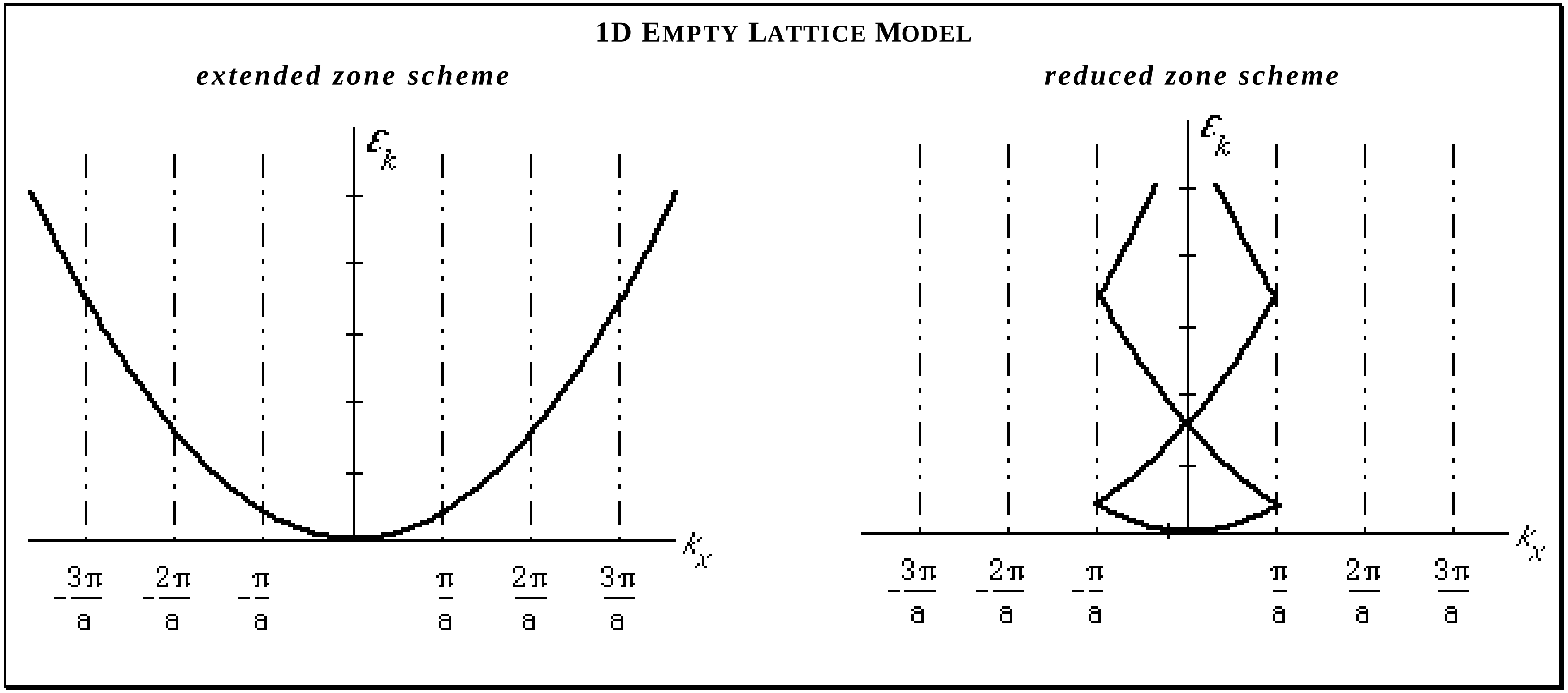
$$\frac{\hbar^2}{2m} \left(\vec{\mathbf{G}}_{[m]} + \vec{\mathbf{k}} \right)^2 u_{[m]}(\vec{\mathbf{k}}) + \sum_{[m']} V_{[m']}\, u_{[m]-[m']}(\vec{\mathbf{k}}) = \varepsilon_{\vec{\mathbf{k}}}\, u_{[m]}(\vec{\mathbf{k}}) \qquad \text{[VII-15]}$$

which can, in principle, be solved for $\varepsilon_{\vec{\mathbf{k}}}$ and $u_{[m]}(\vec{\mathbf{k}})$. If we were to turn off the lattice potential off to effectuate the ***empty lattice approximation***, the eigenfunctions and eigenvalues should, obviously, be those of a **free electron** and given by

$$\psi_{\vec{\mathbf{k}}}(\vec{\mathbf{r}}) \quad \psi^{0}_{\vec{\mathbf{k}}}(\vec{\mathbf{r}}) = (\mathcal{V})^{-1/2} \exp\left(i\, \vec{\mathbf{k}} \quad \vec{\mathbf{r}} \right) \qquad \text{[VII-16]}$$

and

$$\varepsilon_{\vec{\mathbf{k}}} \quad \varepsilon^{0}_{\vec{\mathbf{k}}} = \frac{\hbar^2 \left| \vec{\mathbf{k}} \right|^2}{2m} \qquad \text{[VII-17]}$$



1D EMPTY LATTICE MODEL

extended zone scheme ***reduced zone scheme***

With gradual restoration of the lattice potential, the wave function is gradually transformed from a plane wave, Equation [VII-16], to a Bloch function, Equation [VII-9]. In general, the value of $\vec{\mathbf{k}}$ is well defined since it does not change from its original value and the plane wave merely becomes modulated by the function $u_{\vec{\mathbf{k}}}(\vec{\mathbf{r}})$. Clearly, it is also possible to write the Bloch wave function in the form

$$\psi_{\vec{\mathbf{k}}}(\vec{\mathbf{r}}) = (\mathcal{V})^{-1/2} \left\{ u_{\vec{\mathbf{k}}}(\vec{\mathbf{r}}) \exp\left(i \, \vec{\mathbf{G}}_{[m]} \; \vec{\mathbf{r}} \right) \right\} \exp\left[i \left(\vec{\mathbf{k}} - \vec{\mathbf{G}}_{[m]} \right) \; \vec{\mathbf{r}} \right] \qquad \text{[VII-18]}$$

and since both $u_{\vec{\mathbf{k}}}(\vec{\mathbf{r}})$ and $\exp\left(i \, \vec{\mathbf{G}}_{[m]} \; \vec{\mathbf{r}} \right)$ are spatially periodic, so is their product. Thus, the wave function can be considered as obeying the Bloch theorem with any empty lattice wave vector $\vec{\mathbf{k}} - \vec{\mathbf{G}}_{[m]}$. Accordingly, the theorem allows for two alternative modes of classifying solutions -- *i.e.* a solution may be specified with the original value of $\vec{\mathbf{k}}$ (extended zone scheme) or with that value "reduced" by a reciprocal lattice vector plus a *band index* (reduced zone scheme).

In the ***nearly free electron*** approximation we use the $\psi^0_{\vec{\mathbf{k}}}(\vec{\mathbf{r}})$ functions as a basis set and take the periodic potential $V(\vec{\mathbf{r}})$ as a small static perturbation. Using Equations [A-6c] and [A-7b] of this set of lecture notes (at the end of the Section II, *Review of Basic Quantum Mechanics: Dynamic Behavior of Quantum Systems*) we obtain

$$\left|\varepsilon^1_{\vec{\mathbf{k}}}\right\rangle \approx \left|\varepsilon^0_{\vec{\mathbf{k}}}\right\rangle + \sum \left|\varepsilon^0_{\vec{\mathbf{k}}'}\right\rangle \frac{\left\langle \varepsilon^0_{\vec{\mathbf{k}}'}\right|V\left|\varepsilon^0_{\vec{\mathbf{k}}}\right\rangle}{\varepsilon^0_{\vec{\mathbf{k}}'} - \varepsilon^0_{\vec{\mathbf{k}}}} \qquad \text{[VII-19]}$$

$$\varepsilon_{\vec{\mathbf{k}}} \quad \varepsilon^0_{\vec{\mathbf{k}}} + \sum \frac{\left|\left\langle \varepsilon^0_{\vec{\mathbf{k}}'}\right|V\left|\varepsilon^0_{\vec{\mathbf{k}}}\right\rangle\right|^2}{\varepsilon^0_{\vec{\mathbf{k}}'} - \varepsilon^0_{\vec{\mathbf{k}}}} \qquad \text{[VII-20]}$$

In light of Equation [VII-13a] we may write

$$\begin{aligned} \left\langle \varepsilon^0_{\vec{\mathbf{k}}'}\right|V\left|\varepsilon^0_{\vec{\mathbf{k}}}\right\rangle &= (\mathcal{V})^{-1} \iiint d\vec{\mathbf{r}} \exp\left(-i\,\vec{\mathbf{k}}' \quad \vec{\mathbf{r}}\right) V(\vec{\mathbf{r}}) \exp\left(\,i\,\vec{\mathbf{k}} \quad \vec{\mathbf{r}}\right) \\ &= (\mathcal{V})^{-1} \iiint d\vec{\mathbf{r}} \exp\left[-i\left(\vec{\mathbf{k}}' - \vec{\mathbf{k}}\right) \quad \vec{\mathbf{r}}\right] \sum_{[m]} V_{[m]} \exp\left(\,i\,\vec{\mathbf{G}}_{[m]} \quad \vec{\mathbf{r}}\right) \qquad \text{[VII-21]} \\ &= V_{[m]}\,\delta\left(\vec{\mathbf{k}} + \vec{\mathbf{G}}_{[m]} - \vec{\mathbf{k}}'\right) \end{aligned}$$

so that Equations [VII-19] and [VII-20] become

$$\left|\varepsilon_{\vec{\mathbf{k}}}\right\rangle \approx \left|\varepsilon^0_{\vec{\mathbf{k}}}\right\rangle + \sum_{[m]} \left|\varepsilon^0_{\vec{\mathbf{k}}+\vec{\mathbf{G}}_{[m]}}\right\rangle \frac{V_{[m]}}{\varepsilon^0_{\vec{\mathbf{k}}+\vec{\mathbf{G}}_{[m]}} - \varepsilon^0_{\vec{\mathbf{k}}}} \qquad \text{[VII-22]}$$

$$\varepsilon_{\vec{\mathbf{k}}} \approx \varepsilon^{0}_{\vec{\mathbf{k}}} + \sum_{[m]} \frac{\left| V_{[m]} \right|^{2}}{\varepsilon^{0}_{\vec{\mathbf{k}}+\vec{\mathbf{G}}_{[m]}} - \varepsilon^{0}_{\vec{\mathbf{k}}}} \qquad \text{[VII-23]}$$

Clearly, the strongest departure from free electron behavior occurs when the denominator vanishes -- *i.e.* when

$$\begin{aligned} \varepsilon^{0}_{\vec{\mathbf{k}}+\vec{\mathbf{G}}_{[m]}} - \varepsilon^{0}_{\vec{\mathbf{k}}} &= \frac{\hbar^{2}}{2m} \left(\left| \vec{\mathbf{G}}_{[m]} + \vec{\mathbf{k}} \right|^{2} - \left| \vec{\mathbf{k}} \right|^{2} \right) \\ &= \frac{\hbar^{2}}{2m} \left(\left| \vec{\mathbf{G}}_{[m]} \right|^{2} + 2\vec{\mathbf{G}}_{[m]} \cdot \vec{\mathbf{k}} \right) = 0 \end{aligned} . \qquad \text{[VII-24]}$$

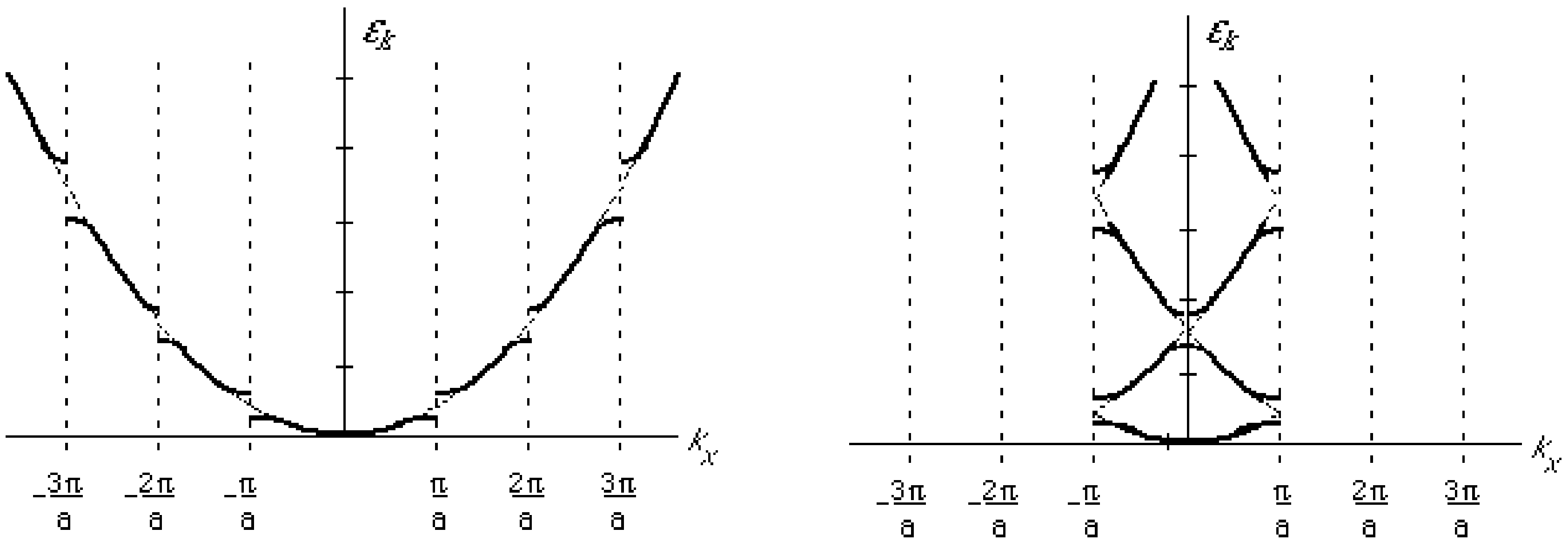
Equation[VII-24] is the famous **Bragg scattering condition** or the condition which defines the boundaries of the Brillouin zones. At the zone edge for a particular pair of degenerate free electron states the appropriate pair of equation from the set in Equation [VII-19] may be approximated

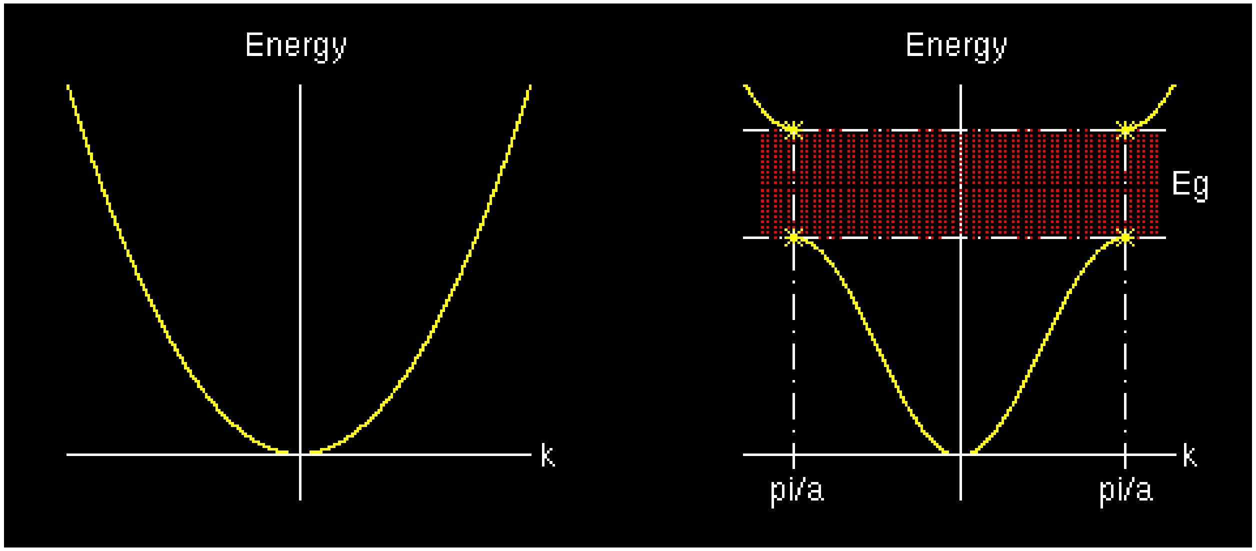
$$\begin{aligned} \left[\varepsilon^{0}_{\vec{\mathbf{k}}} - \varepsilon_{\vec{\mathbf{k}}} \right] u_{[0]}\left(\vec{\mathbf{k}}\right) + V_{[m]}\, u_{[m]}\left(\vec{\mathbf{k}}\right) &= 0 \\ \left[\varepsilon^{0}_{\vec{\mathbf{k}}+\vec{\mathbf{G}}_{[m]}} - \varepsilon_{\vec{\mathbf{k}}} \right] u_{[m]}\left(\vec{\mathbf{k}}\right) + V_{[m]}\, u_{[0]}\left(\vec{\mathbf{k}}\right) &= 0 \end{aligned} \qquad \text{[VII-25a]}$$

or

$$\varepsilon_{\vec{\mathbf{k}}} = \frac{1}{2} \left\{ \varepsilon^{0}_{\vec{\mathbf{k}}+\vec{\mathbf{G}}_{[m]}} + \varepsilon^{0}_{\vec{\mathbf{k}}} \pm \sqrt{\left(\varepsilon^{0}_{\vec{\mathbf{k}}+\vec{\mathbf{G}}_{[m]}} - \varepsilon^{0}_{\vec{\mathbf{k}}} \right)^{2} + 4\left| V_{[m]} \right|^{2}} \right\} . \qquad \text{[VII-25b]}$$

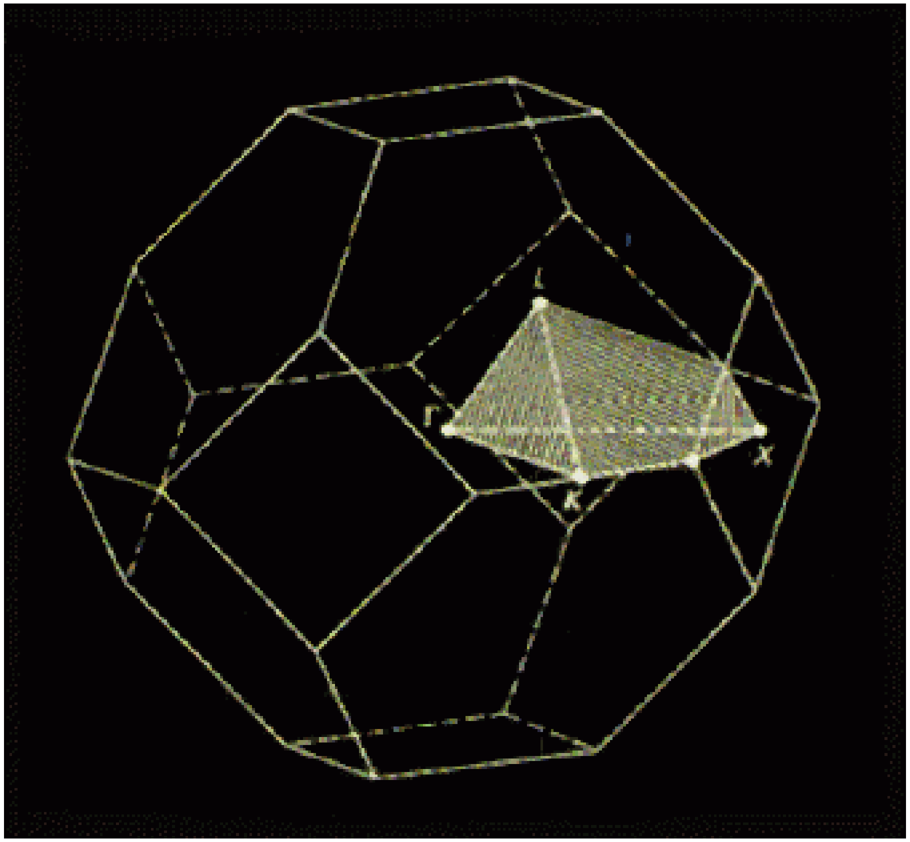
1D NEARLY FREE ELECTRON MODEL

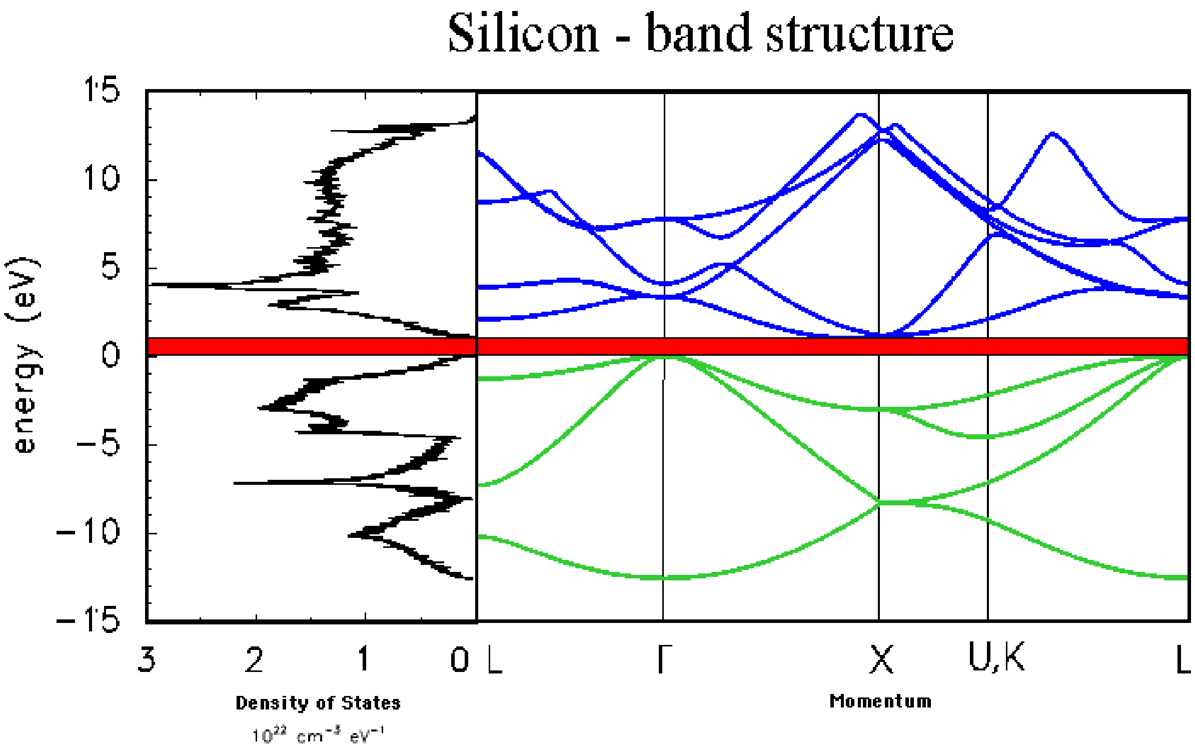

extended zone scheme *reduced zone scheme*



These considerations then provide the context for the examination of ***real*** band structures.

Diamond Structure Brillouin Zone and Nomincature





For more on band structures go to:

http://www.deas.harvard.edu/courses/ap216/images/bandgap_engineering/bandgap_engineering.html

For more on pn junction physics go to:

http://www.deas.harvard.edu/courses/ap216/images/pn_junction/pn_junction.html

B. OPTICAL PROPERTIES OF BULK (3-D) SEMICONDUCTOR[32]

FREE-CARRIER THEORY

For most of our considerations a **parabolic-band model** provides a reasonable picture of optical properties. In this model we assume that near a band edge the single particle energy of the electron **measured with respect to the edge** is given by

$$\varepsilon_{\vec{\mathbf{k}}} = \frac{\hbar^2 \left|\vec{\mathbf{k}}\right|^2}{2m} = \frac{\hbar^2 k^2}{2m} \qquad \text{[VII-26a]}$$

where the ***effective mass*** m is a measure of the inverse curvature of the band.[33] Introducing the notion of a deficiency of electrons in the valence band -- *i.e.* **holes** -- as positive charge carriers, the parabolic-band model is re-interpreted as, respectively, the quasiparticle energies of an electron in the conduction band and a hole in the valence band

$$\varepsilon_{e,\vec{\mathbf{k}}} = \frac{\hbar^2 \left|\vec{\mathbf{k}}\right|^2}{2m_e} = \frac{\hbar^2 k^2}{2m_e} \qquad \text{[VII-26b]}$$

$$\varepsilon_{h,\vec{\mathbf{k}}} = \frac{\hbar^2 \left|\vec{\mathbf{k}}\right|^2}{2m_h} = \frac{\hbar^2 k^2}{2m_h} \qquad \text{[VII-26c]}$$

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[32] This section draws heavily on Weng W. Chow, Stephan W. Koch and Murray. Sargent III, *Semiconductor-Laser Physics*, Springer-Verlag (1994)

[33] That is, the effective mass is given by

$$\frac{1}{m} = \frac{1}{\hbar^2}\frac{\partial^2}{\partial k^2}\varepsilon_{\vec{\mathbf{k}}}\Big|_{\text{Band edge}}$$

and, thus, in the **conduction band the effective mass of electrons is positive and in the valence band it is negative** -- *i.e.* $m_c > 0$ and $m_v < 0$.

where $m_e = m_c > 0$ and $m_h > 0$ accounts for aggregate dynamics of all the electron in the valence minus the single empty state.[34] Of course, the single particle picture discussed thus far, does not include the critically important effects of the inter-electronic Coulomb interactions and, in particular, ***carrier-carrier scattering***. In what is usually called ***free-carrier theory***, it is assumed that carrier-carrier scattering causes a rapid "thermalization" (relaxation times less than 0.1 picoseconds) of excited conduction band electrons (and valence band holes) and, consequently, it is assumed that carriers **within a band** are in *quasiequilibrium* with energies distributed according to a Fermi-Dirac distribution (see illustration on the next page).[35],, [36] Accordingly, the carrier density for a given band (*i.e.* $\alpha = e$ for the conduction band and $\alpha = h$ for the valence band) is determined by the condition

$$N_\alpha = \frac{1}{V}\sum_{\text{states}} f_{\alpha\vec{k}} = \frac{1}{V}\sum_{\text{states}} \left\{\exp\left[\beta\left(\varepsilon_{\alpha\vec{k}} - \mu_\alpha\right)\right]+1\right\}^{-1} \qquad \text{[VII-27]}$$

where V is the volume of the sample and μ_α is the carrier ***quasichemical potential*** (or ***imref*** ≡ **quasi-Fermi energy**).[37] In the discussion of Equation

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[34] The hole energy is the energy of the filled valence minus the energy of the filled with a vacant electronic state so that an increase in hole momentum leads to an increase in hole energy. Since m_h must of necessity include the many-body Coulomb interactions among the valence electrons.

[35] See a Derivation of the Fermi-Dirac distribution function at http://ece-www.colorado.edu/~bart/book/fermi-dirac_derivation.htm

[36] See the applets Fermi Level vs. Carrier Concentration and Doping of Donor and Acceptor Impurities
(at http://www.acsu.buffalo.edu/~wie/applet/fermi/fermi.html)
and Fermi Level, Fermi Function and Electron Occupancy of Localized Energy States
(at http://www.acsu.buffalo.edu/~wie/applet/fermi/functionAndStates/functionAndStates.html).

[37] As are the electron and hole energies $\varepsilon_{\alpha\vec{k}}$, the imrefs μ_α are measured with respect to the appropriate band edge -- *viz.*

$$\mu_e = \left(E_F\right)_C - E_C \quad \text{and} \quad \mu_h = E_V - \left(E_F\right)_V$$

Thus, if $\mu_\alpha < 0$ the band does contain enough carriers to populate any state with a probability greater than one-half.

[IV-5] of this set of lectures, it was argued that the 3-D density of states in $\vec{\mathbf{k}}$ space is given by

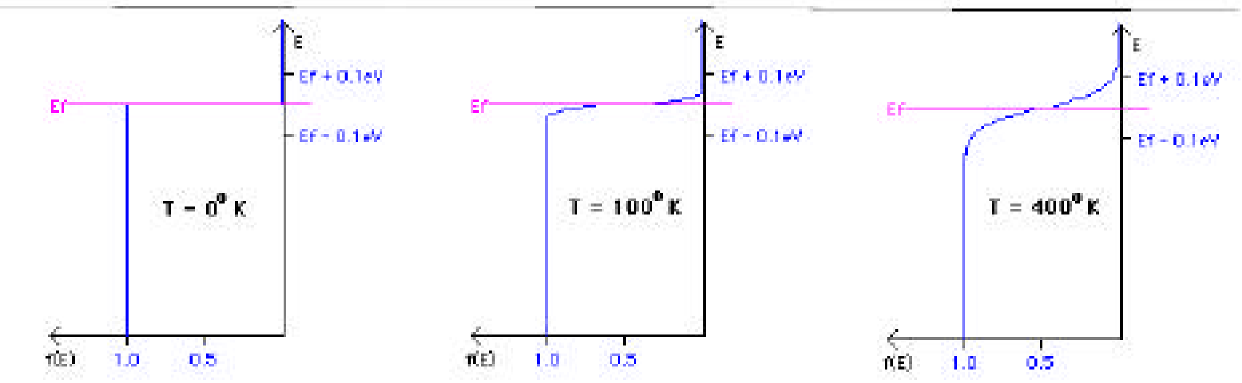
$$\rho(k)\;dk = \frac{\mathrm{V}}{\pi^2}\,k^2\;dk \qquad [\text{ VII-28 }]$$

which translates (within a given band) into a density of states per unit energy per unit volume of

$$\rho\left(\varepsilon_{\alpha\vec{\mathbf{k}}}\right) = \frac{1}{\mathrm{V}}\,\rho(k)\,\frac{dk}{d\varepsilon_{\alpha\vec{\mathbf{k}}}} = \frac{1}{2\pi^2}\left(\frac{2m_\alpha}{\hbar^2}\right)^{3/2}\sqrt{\varepsilon_{\alpha\vec{\mathbf{k}}}} \qquad [\text{ VII-29}]$$

Hence, for a 3D semiconductor, Equation [VII-27] becomes

$$N_\alpha = \int_0^\infty \rho\left(\varepsilon_\alpha\right)\,f\left(\varepsilon_\alpha\right)\,d\varepsilon_\alpha = \frac{1}{2\pi^2}\left(\frac{2m_\alpha}{\hbar^2}\right)^{3/2}\int_0^\infty \sqrt{\varepsilon_\alpha}\,\left\{\exp\left[\,\beta\left(\varepsilon_\alpha - \mu_\alpha\right)\right]\right\}^{-1}\,d\varepsilon_\alpha \qquad [\text{ VII-30 }]$$


Fermi Function

OPTICAL MATRIX ELEMENTS

Since optical interactions in a semiconductor are essential **distributed**, it is probably more precise and appropriate in this case to write the interaction Hamiltonian as [38]

$$\mathcal{H}_{\mathrm{I}} = -\frac{q}{m}\vec{\mathbf{A}} \cdot \vec{\mathcal{P}} = \frac{e}{m}\vec{\mathbf{A}} \cdot \vec{\mathcal{P}} \qquad \frac{e\,A_0}{2m}\left[\hat{e} \cdot \vec{\mathcal{P}}\right]\exp\left[\,i\left(\omega_{\mathrm{rad}}\,t - \vec{\mathbf{k}}_{\mathrm{rad}} \cdot \vec{\mathbf{r}}\right)\right] + \ldots \qquad \text{[VII-31]}$$

Thus, the critical optical matrix element [39] between a state in the valence band and one in the conduction band is

$$\left\langle \mathcal{E}_{\mathrm{v}\,\vec{\mathbf{k}}}\right| \mathcal{H}_{\mathrm{I}} \left|\mathcal{E}_{\mathrm{c}\,\vec{\mathbf{k}}'}\right\rangle = \frac{eA_0}{2m}\exp\left[\,i\,\omega_{\mathrm{rad}}t\right]\overline{\overline{\mathbf{M}}}_{\mathrm{v}\,\vec{\mathbf{k}},\mathrm{c}\,\vec{\mathbf{k}}'} \qquad \text{[VII-32a]}$$

where the so called ***transition matrix element*** is given by

$$\overline{\overline{\mathbf{M}}}_{\mathrm{v}\,\vec{\mathbf{k}},\mathrm{c}\,\vec{\mathbf{k}}'} \equiv \int d\vec{\mathbf{r}}\; u_{\mathrm{v}\,\vec{\mathbf{k}}}\left(\vec{\mathbf{r}}\right)\exp\left[-i\,\vec{\mathbf{k}} \cdot \vec{\mathbf{r}}\right]\left\{\hat{e} \cdot \vec{\mathcal{P}}\,\exp\left[-i\,\vec{\mathbf{k}}_{\mathrm{rad}} \cdot \vec{\mathbf{r}}\right]\right\} u_{\mathrm{c}\,\vec{\mathbf{k}}'}\left(\vec{\mathbf{r}}\right)\exp\left[\,i\,\vec{\mathbf{k}}' \cdot \vec{\mathbf{r}}\right] \qquad \text{[VII-32b]}$$

Using the expansion for $u_{\mathrm{n}\,\vec{\mathbf{k}}}$ in Equation [VII-12a] the matrix element becomes

$$\begin{aligned}\overline{\overline{\mathbf{M}}}_{\mathrm{v}\,\vec{\mathbf{k}},\,\mathrm{c}\,\vec{\mathbf{k}}'} &= \sum_{[m]}\sum_{[m']} u_{\mathrm{v}[m]}\left(\vec{\mathbf{k}}\right) u_{\mathrm{c}\,[m']}\left(\vec{\mathbf{k}}'\right) \\ &\quad \times \iiint d\vec{\mathbf{r}}\,\exp\left[-i\left(\vec{\mathbf{k}} + \vec{\mathbf{k}}_{\mathrm{rad}} + \vec{\mathbf{G}}_{[m]}\right) \cdot \vec{\mathbf{r}}\right]\left\{\hat{e} \cdot \vec{\mathcal{P}}\right\}\exp\left[\,i\left(\vec{\mathbf{k}}' + \vec{\mathbf{G}}_{[m']}\right) \cdot \vec{\mathbf{r}}\right]\end{aligned} \qquad \text{[VII-33]}$$

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[38] In the next set of lectures, *The Interaction of Radiation and Matter: Quantum Theory,* we develop the following expression for the **nonrelativistic Hamiltonian of a single charged particle**:

$$\mathcal{H} = q\phi + \frac{1}{2m}\left[\,\vec{\mathcal{P}} - q\,\vec{\mathbf{A}}\,\right]^2$$

where $\vec{\mathcal{P}}$ is the canonical conjugate momentum of the charged particle.

[39] The Fermi Golden Rule and first-order perturbation theory tells us that the **radiation-induced transition rate** is given by

$$\sum_f \frac{2\pi}{\hbar}\left|\langle\, f \,|\, \mathcal{H}_{\mathrm{int}} \,|\, i\,\rangle\right|\delta\!\left(\mathcal{E}_f - \mathcal{E}_i\right) .$$

However, $\vec{\mathcal{P}} \quad -i\hbar\,\vec{}$ so that

$$\overline{\overline{\mathbf{M}}}_{\mathrm{v}\,\vec{\mathbf{k}},\mathrm{c}\,\vec{\mathbf{k}}'} = \sum_{[m]}\sum_{[m']}\left\{\hbar\,\hat{e}\;\left(\vec{\mathbf{k}}'+\vec{\mathbf{G}}_{[m']}\right)\right\} u_{\mathrm{v}\,[m]}\!\left(\vec{\mathbf{k}}\right) u_{\mathrm{c}\,[m']}\!\left(\vec{\mathbf{k}}'\right) \times \iiint d\vec{\mathbf{r}}\,\exp\left[-i\left(\vec{\mathbf{k}}+\vec{\mathbf{k}}_{\mathrm{rad}}-\vec{\mathbf{k}}'+\vec{\mathbf{G}}_{[m]}-\vec{\mathbf{G}}_{[m']}\right)\;\vec{\mathbf{r}}\right] \qquad \text{[VII-34]}$$

With the parsing $\vec{\mathbf{r}} = \vec{\mathbf{R}}_{[\ell]} + \vec{\rho}$ and using the defining relationship $\exp\left(\,i\,\vec{\mathbf{R}}_{[\ell]}\quad\vec{\mathbf{G}}_{[m]}\right) = 1$, the volume integral may be expressed

$$\iiint d\vec{\mathbf{r}}\left\{\vec{\mathbf{r}}\right\}\,\exp\left[\;i\left(\vec{\mathbf{k}}'-\vec{\mathbf{k}}-\vec{\mathbf{k}}_{\mathrm{rad}}+\vec{\mathbf{G}}_{[m']}-\vec{\mathbf{G}}_{[m]}\right)\;\vec{\mathbf{r}}\right]$$

$$= \iiint_{\text{unit cell}} d\vec{\mathbf{r}}\left\{\vec{\rho}\right\}\,\exp\left[\;i\left(\vec{\mathbf{k}}'-\vec{\mathbf{k}}-\vec{\mathbf{k}}_{\mathrm{rad}}+\vec{\mathbf{G}}_{[m']}-\vec{\mathbf{G}}_{[m]}\right)\;\vec{\rho}\right] \times \sum_{[\ell]}\exp\left[\;i\left(\vec{\mathbf{k}}'-\vec{\mathbf{k}}-\vec{\mathbf{k}}_{\mathrm{rad}}\right)\;\vec{\mathbf{R}}_{[\ell]}\right] \qquad \text{[VII-35]}$$

The summation $\sum_{[\ell]}\exp\left[\;i\left(\vec{\mathbf{k}}'-\vec{\mathbf{k}}-\vec{\mathbf{k}}_{\mathrm{rad}}\right)\;\vec{\mathbf{R}}_{[\ell]}\right] \qquad 0$ unless

$$\vec{\mathbf{k}}' = \vec{\mathbf{k}} + \vec{\mathbf{k}}_{\mathrm{rad}} \approx \vec{\mathbf{k}}. \qquad \text{[VII-36]}$$

-- *i.e.* essentially **only vertical transitions are allowed**! Therefore,

$$\overline{\overline{\mathbf{M}}}_{\mathrm{v}\,\vec{\mathbf{k}},\mathrm{c}\,\vec{\mathbf{k}}'} \qquad \delta_{\vec{\mathbf{k}}\,\vec{\mathbf{k}}'}\left(\frac{V_{\text{sample}}}{V_{\text{unit cell}}}\right)\sum_{[m]}\sum_{[m']}\left\{\hbar\,\hat{e}\;\left(\vec{\mathbf{k}}+\vec{\mathbf{G}}_{[m']}\right)\right\} u_{\mathrm{v}\,[m]}\!\left(\vec{\mathbf{k}}\right) u_{\mathrm{c}\,[m']}\!\left(\vec{\mathbf{k}}\right) \times \iiint_{\text{unit cell}} d\vec{\mathbf{r}}\,\exp\left[\;i\left(\vec{\mathbf{G}}_{[m']}-\vec{\mathbf{G}}_{[m]}\right)\;\vec{\rho}\right] \qquad .\text{[VII-37a]}$$

For a given polarization of the applied field, the transition matrix can be written

$$\overline{\overline{\mathbf{M}}}_{v\vec{\mathbf{k}}c\,\vec{\mathbf{k}}'} = \left\{\text{polarization factor}\right\} \overline{\overline{\mathbf{M}}} \; . \qquad [\text{ VII-37b }]$$

where the polarization factors are of order one and typical values of $|\mathbf{M}|^2$ are given in the following table:[40]

Material system	$2|\mathbf{M}|^2/m$ (in eV)
Ga As	28.8
$Al_x\,Ga_{1-x}\,As$ (x < 0.3)	29.83 + 2.85 x
$In_x\,Ga_{1-x}\,As$	28.8 - 6.6 x
In P	19.7
$In_{1-x}\,Ga_x\,As_y\,P_{1-y}$ (x = 0.47 y)	19.7 + 5.6 y

Of course, the radiation must also satisfy the energy conservation condition

$$\Delta\mathcal{E}_{\vec{\mathbf{k}}} = \hbar\,\omega_{\vec{\mathbf{k}}} = \mathcal{E}_g + \frac{\hbar^2\,k^2}{2m_e} + \frac{\hbar^2\,k^2}{2\,m_h} = \mathcal{E}_g + \frac{\hbar^2\,k^2}{2m_r} \qquad [\text{ VII-38a }]$$

where

$$\frac{1}{m_r} = \frac{1}{m_e} + \frac{1}{m_h} \; . \qquad [\text{ VII-38b }]$$

"FREE-CARRIER THEORY" OF OPTICAL PROCESSESS

The *bottom line* is that in the ***free-carrier theory*** of optical interactions, the effective Hamiltonian for the carriers -- *i.e.* "free" particle kinetic energy plus electromagnetic interaction -- is separable into a series of $\vec{\mathbf{k}}$ -dependent terms.[41] Thus, if we neglect $\vec{\mathbf{k}}\vec{\mathbf{k}}'$ correlations in the treatment of optical properties, we need only consider the following manifold of states:

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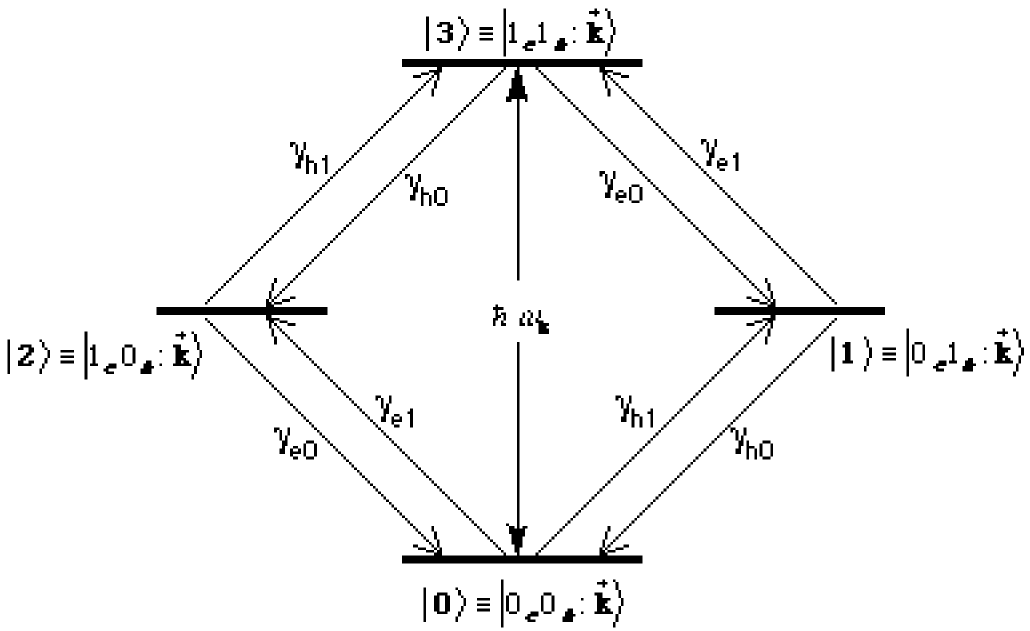
[40] From *Diode Lasers and Photonic Integrated Circuits* by Larry A. Coldren and Scott W. Corzine, Wiley (1995)

[41] In particular, it can be shown -- see Weng W. Chow, Stephan W. Koch and Murray. Sargent III, *Semiconductor-Laser Physics*, Springer-Verlag (1994) -- that

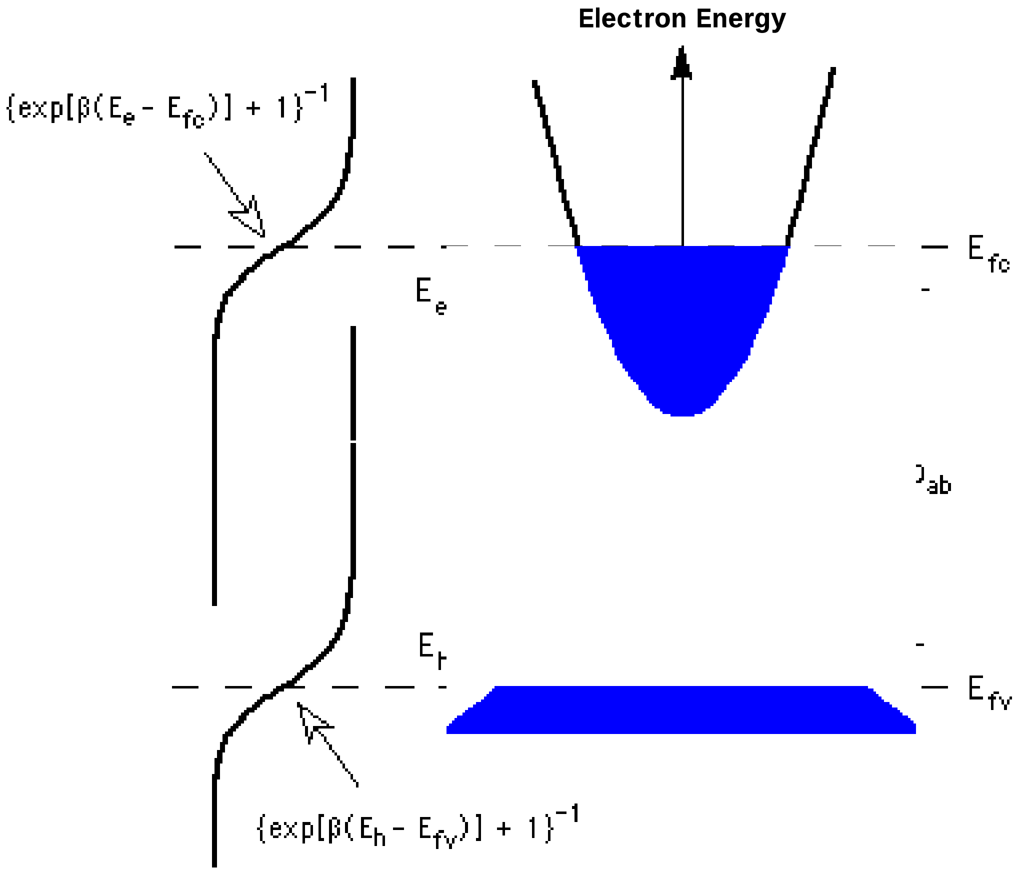

$|3\rangle \equiv |1_e 1_h : \vec{\mathbf{k}}\rangle$
γ_{h1}
γ_{h0}
γ_{e0}
γ_{e1}
$|2\rangle \equiv |1_e 0_h : \vec{\mathbf{k}}\rangle$
$\hbar\,\omega_{\vec{\mathbf{k}}}$
$|1\rangle \equiv |0_e 1_h : \vec{\mathbf{k}}\rangle$
γ_{e0}
γ_{e1}
γ_{h1}
γ_{h0}
$|0\rangle \equiv |0_e 0_h : \vec{\mathbf{k}}\rangle$

Equation [VII-37a] tells us that the absorption or emission of a single photon connect only the two states in the manifold which can be identified as $\left|0_e\ 0_h : \vec{\mathbf{k}}\right\rangle$ and $\left|1_e\ 1_h : \vec{\mathbf{k}}\right\rangle$ where the first number in the ket specifies **electron occupancy** of a state with momentum $\hbar\,\vec{\mathbf{k}}$ and the second **hole occupancy** of a state with momentum $-\hbar\,\vec{\mathbf{k}}$.

$$\mathcal{H}_{\text{eff}} = \sum_{\mathbf{k}} \left\{ \left[\mathcal{E}_g + \frac{\hbar^2\ k^2}{2m_{ec}} \right] a_{\vec{\mathbf{k}}}^{\dagger}\ a_{\vec{\mathbf{k}}} + \left[\frac{\hbar^2\ k^2}{2m_{hv}} \right] b_{-\vec{\mathbf{k}}}^{\dagger}\ b_{-\vec{\mathbf{k}}} - \left[\mu_{\vec{\mathbf{k}}}\ a_{\vec{\mathbf{k}}}^{\dagger}\ b_{-\vec{\mathbf{k}}}^{\dagger} + \mu_{\vec{\mathbf{k}}}\ a_{\vec{\mathbf{k}}}\ b_{-\vec{\mathbf{k}}} \right] \mathrm{E}(z,t) \right\}$$

where $\left\{a_{\vec{\mathbf{k}}}^{\dagger}, a_{\vec{\mathbf{k}}}\right\}$ and $\left\{b_{\vec{\mathbf{k}}}^{\dagger}, b_{\vec{\mathbf{k}}}\right\}$ are, respectively, electron and hole {**creation**, **destruction**} operators. $\mu_{\vec{\mathbf{k}}}$ is the dipole matrix element between vertical states in the valence and conduction bands.

ENERGY DIAGRAM FOR BULK LASERS


Electron Energy
$\{\exp[\beta(E_e - E_{fc})] + 1\}^{-1}$
E_{fc}
E_e
ℷab
E_h
E_{fv}
$\{\exp[\beta(E_h - E_{fv})] + 1\}^{-1}$

Since no $\vec{\mathbf{k}}\vec{\mathbf{k}}'$ correlations are involved in the free carrier theory, the density operator of the complete system may be expressed as a product of component density operators

$$\rho = \prod_{\vec{\mathbf{k}}} \rho\left(\vec{\mathbf{k}}\right) \qquad \text{[VII-39]}$$

so that the Schrödinger equation of motion for the density operator is also separable -- *viz*.

$$\dot{\rho}\left(\vec{\mathbf{k}}\right) = -\frac{i}{\hbar}\left[\, \mathcal{H}_{\text{eff}}\left(\vec{\mathbf{k}}\right), \rho\left(\vec{\mathbf{k}}\right) \right] . \qquad [\text{ VII-39 }]$$

C. INJECTION LASER THEORY

(For pictorials on laser configurations go to:
http://www.deas.harvard.edu/courses/ap216/images/injection_lasers/laser_structures/laser_structures.html)

A fairly satisfactory model of lasing (and other optical processes) in semiconductors may be obtain by adaptation of the two-level, semiclassical discussed earlier. In light of the discussion in the previous section, we adapt the two-level theory by making the identifications

$$\begin{array}{ll}
\rho_{aa}\left(\vec{\mathbf{k}}\right) & \rho_{1111}\left(\vec{\mathbf{k}}\right) = \left\langle 1_e 1_h : \vec{\mathbf{k}} \left| \rho \right| 1_e 1_h : \vec{\mathbf{k}} \right\rangle = \left\langle 3:\vec{\mathbf{k}} \left| \rho \right| 3:\vec{\mathbf{k}} \right\rangle = \rho_{33}\left(\vec{\mathbf{k}}\right) \\
\rho_{bb}\left(\vec{\mathbf{k}}\right) & \rho_{00\ 00}\left(\vec{\mathbf{k}}\right) = \left\langle 0_e\ 0_h : \vec{\mathbf{k}} \left| \rho \right| 0_e\ 0_h : \vec{\mathbf{k}} \right\rangle = \left\langle 0:\vec{\mathbf{k}} \left| \rho \right| 0:\vec{\mathbf{k}} \right\rangle = \rho_{00}\left(\vec{\mathbf{k}}\right) \\
\rho_{ab}\left(\vec{\mathbf{k}}\right) & \rho_{1100}\left(\vec{\mathbf{k}}\right) = \left\langle 1_e 1_h : \vec{\mathbf{k}} \left| \rho \right| 0_e\ 0_h : \vec{\mathbf{k}} \right\rangle = \left\langle 3:\vec{\mathbf{k}} \left| \rho \right| 0:\vec{\mathbf{k}} \right\rangle == \rho_{30}\left(\vec{\mathbf{k}}\right)
\end{array} \qquad [\text{ VII--40 }]$$

Given the **free carrier effective Hamiltonian** discussed above and the equation of motion expressed in Equation [VII-31], we can write

$$\begin{aligned}
\dot{\rho}_{30}\left(\vec{\mathbf{k}}\right) &= \left\langle 3:\vec{\mathbf{k}} \left| \dot{\rho}\left(\vec{\mathbf{k}}\right) \right| 0:\vec{\mathbf{k}} \right\rangle \\
&= -\frac{i}{\hbar}\left[\left\langle 3:\vec{\mathbf{k}} \left| \mathcal{H}_{\text{eff}}\left(\vec{\mathbf{k}}\right) \right| 3:\vec{\mathbf{k}} \right\rangle - \left\langle 0:\vec{\mathbf{k}} \left| \mathcal{H}_{\text{eff}}\left(\vec{\mathbf{k}}\right) \right| 0:\vec{\mathbf{k}} \right\rangle\right] \rho_{30}\left(\vec{\mathbf{k}}\right) \\
&\qquad + \frac{i}{\hbar}\left\langle 3:\vec{\mathbf{k}} \left| \mathcal{H}_{\text{eff}}\left(\vec{\mathbf{k}}\right) \right| 0:\vec{\mathbf{k}} \right\rangle \left[\rho_{33}\left(\vec{\mathbf{k}}\right) - \rho_{00}\left(\vec{\mathbf{k}}\right)\right]
\end{aligned} \qquad [\text{ VII--41a }]$$

$$\begin{aligned}
\dot{\rho}_{33}\left(\vec{\mathbf{k}}\right) = \left\langle 3:\vec{\mathbf{k}} \left| \dot{\rho}\left(\vec{\mathbf{k}}\right) \right| 3:\vec{\mathbf{k}} \right\rangle &= \frac{i}{\hbar}\left[\rho_{30}\left(\vec{\mathbf{k}}\right)\left\langle 0:\vec{\mathbf{k}} \left| \mathcal{H}_{\text{eff}}\left(\vec{\mathbf{k}}\right) \right| 3:\vec{\mathbf{k}} \right\rangle\right] \\
&\qquad - \frac{i}{\hbar}\left[\rho_{03}\left(\vec{\mathbf{k}}\right)\left\langle 3:\vec{\mathbf{k}} \left| \mathcal{H}_{\text{eff}}\left(\vec{\mathbf{k}}\right) \right| 0:\vec{\mathbf{k}} \right\rangle\right]
\end{aligned} \qquad [\text{ VII--41b }]$$

$$\rho_{00}\left(\vec{\mathbf{k}}\right)=\left\langle\, 0{:}\vec{\mathbf{k}}\,\middle|\rho\left(\vec{\mathbf{k}}\right)\middle|\, 0{:}\vec{\mathbf{k}}\,\right\rangle=-\frac{i}{\hbar}\left[\,\rho_{30}\left(\vec{\mathbf{k}}\right)\left\langle\, 0{:}\vec{\mathbf{k}}\,\middle|\,\mathcal{H}_{\text{eff}}\left(\vec{\mathbf{k}}\right)\middle|\, 3{:}\vec{\mathbf{k}}\,\right\rangle\right] \\ +\frac{i}{\hbar}\left[\,\rho_{03}\left(\vec{\mathbf{k}}\right)\left\langle\, 3{:}\vec{\mathbf{k}}\,\middle|\,\mathcal{H}_{\text{eff}}\left(\vec{\mathbf{k}}\right)\middle|\, 0{:}\vec{\mathbf{k}}\,\right\rangle\right] \qquad \text{[VII--41c]}$$

which tells us, once again, that $\rho_{ab}\quad\rho_{30}$ is driven by $\rho_{aa}-\rho_{bb}\quad\rho_{33}-\rho_{00}$ and *vice versa*. There is, however, an important new element to be considered in this problem to wit, as we have illustrated above, we are, in fact, actually dealing with a **four-level system** which includes of the states $\left|\,1_e\,0_h{:}\vec{\mathbf{k}}\,\right\rangle$ and $\left|\,0_e\,1_h{:}\vec{\mathbf{k}}\,\right\rangle$ as well as the pseudo "a" and "b" states. Fortunately, inclusion of these states does not unduly complicate the analysis since, as discussed above, electron-electron scattering induces a rapid relaxation or "thermalization" of the probability of finding a given state occupied. After thermalization, the probability of finding a state with a particular momentum value in a given band is provided by a Fermi-Dirac distribution referenced to a quasi-Fermi energy or **imref** appropriate to that band. To obtain an expression for $\rho_{\text{aa}}-\rho_{\text{bb}}$ we note that[42]

$$\operatorname{tr}\left[\,\rho\left(\vec{\mathbf{k}}\right)\,\right]=\rho_{00}\left(\vec{\mathbf{k}}\right)+\rho_{11}\left(\vec{\mathbf{k}}\right)+\rho_{22}\left(\vec{\mathbf{k}}\right)+\rho_{33}\left(\vec{\mathbf{k}}\right)=1 \qquad \text{[VII--42]}$$

so that

$$\begin{aligned}\rho_{\text{aa}}-\rho_{\text{bb}}&=\rho_{33}\left(\vec{\mathbf{k}}\right)-\rho_{00}\left(\vec{\mathbf{k}}\right)=\left\{\,\rho_{33}\left(\vec{\mathbf{k}}\right)+\rho_{22}\left(\vec{\mathbf{k}}\right)\right\}+\left\{\,\rho_{33}\left(\vec{\mathbf{k}}\right)+\rho_{11}\left(\vec{\mathbf{k}}\right)\right\}-1\\ &=n_e\left(\vec{\mathbf{k}}\right)+n_h\left(\vec{\mathbf{k}}\right)-1\end{aligned} \qquad \text{[VII--43]}$$

where $n_e\left(\vec{\mathbf{k}}\right)$ is the probability[43] of finding a electron with momentum $\hbar\,\vec{\mathbf{k}}$ independent of whether or not there is a hole with momentum $-\hbar\,\vec{\mathbf{k}}$ and $n_h\left(\vec{\mathbf{k}}\right)$ is the corresponding probability for a hole. Adapting Equations [VI-13a-c] from earlier in this set of lecture notes,[44] we may write

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[42] That is, there must be either 0 or 1 electrons and 0 or 1 holes in the given $\vec{\mathbf{k}}$ state.

[43] In the second quantization representation of the problem, the n's are expectation values of the appropriate number operators.

$$\dot{n}_e\left(\vec{\mathbf{k}}\right) = \Lambda_{e\vec{\mathbf{k}}} - \gamma_{\mathrm{nr}}\, n_e\left(\vec{\mathbf{k}}\right) - \Gamma_{\vec{\mathbf{k}}}\, n_e\left(\vec{\mathbf{k}}\right) n_h\left(\vec{\mathbf{k}}\right) - \dot{n}_e\left(\vec{\mathbf{k}}\right)\Big|_{\text{c-c}} - \left[\, i\,\hbar^{-1}\, \mathcal{V}_{\mathrm{ab}}\left(z,t;\vec{\mathbf{k}}\right) \rho_{\mathrm{ab}}\left(\vec{\mathbf{k}}\right) + c.c \right] \qquad [\text{ VII--44a }]$$

$$\dot{n}_h\left(\vec{\mathbf{k}}\right) = \Lambda_{h\vec{\mathbf{k}}} - \gamma_{\mathrm{nr}}\, n_h\left(\vec{\mathbf{k}}\right) - \Gamma_{\vec{\mathbf{k}}}\, n_e\left(\vec{\mathbf{k}}\right) n_h\left(\vec{\mathbf{k}}\right) - \dot{n}_h\left(\vec{\mathbf{k}}\right)\Big|_{\text{c-c}} - \left[i\,\hbar^{-1}\, \mathcal{V}_{\mathrm{ab}}\left(z,t;\vec{\mathbf{k}}\right) \rho_{\mathrm{ab}}\left(\vec{\mathbf{k}}\right) + c.c \right] \qquad [\text{ VII--44b }]$$

$$\dot{\rho}_{\mathrm{ab}}\left(\vec{\mathbf{k}}\right) = -\left(i\,\omega_{\mathrm{ab}} + \gamma_{\mathrm{ab}}\right) \rho_{\mathrm{ab}}\left(\vec{\mathbf{k}}\right) + i\,\hbar^{-1}\, \mathcal{V}_{\mathrm{ab}}\left(z,t;\vec{\mathbf{k}}\right) \left[n_e\left(\vec{\mathbf{k}}\right) + n_h\left(\vec{\mathbf{k}}\right) - 1 \right] \qquad [\text{ VII--44c }]$$

where $\Lambda_{\alpha\vec{\mathbf{k}}}$ ($\alpha = e$ or h) represents the pumping rate due to carrier injection and $\dot{n}_\alpha\left(\vec{\mathbf{k}}\right)\Big|_{\text{c-c}}$ the effect of carrier-carrier scattering. $\gamma_{\mathrm{nr}}\, n_\ell\left(\vec{\mathbf{k}}\right)$ and $\Gamma_{\vec{\mathbf{k}}}\, n_e\left(\vec{\mathbf{k}}\right) n_h\left(\vec{\mathbf{k}}\right)$ are, respectively, phenomenological representations of nonradiative decay and radiative "bimolecular" recombination (**spontaneous emission**), respectively. The probability difference (**gain factor)**

$$\Delta\left(\vec{\mathbf{k}}\right) \equiv n_e\left(\vec{\mathbf{k}}\right) + n_h\left(\vec{\mathbf{k}}\right) - 1 = n_e\left(\vec{\mathbf{k}}\right) n_h\left(\vec{\mathbf{k}}\right) \left\{ 1 - \left[\frac{1}{n_e\left(\vec{\mathbf{k}}\right)} - 1 \right] \left[\frac{1}{n_h\left(\vec{\mathbf{k}}\right)} - 1 \right] \right\} \qquad [\text{ VII--45 }]$$

is the **critical factor** in the analysis of **stimulated** processes -- *i.e.* for inversion $\Delta\left(\vec{\mathbf{k}}\right) > 0$. This gain factor may be written as $\Delta\left(\vec{\mathbf{k}}\right) = n_e\left(\vec{\mathbf{k}}\right) - \left[1 - n_h\left(\vec{\mathbf{k}}\right)\right]$ -- *i.e.* the population inversion is proportional to the probability difference of an electron in corresponding $\vec{\mathbf{k}}$-states in the conduction and valence bands. Alternatively and more usefully, we see that it varies directly with the product of

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[44] That is

$$\dot{\bar{\rho}}_{\mathrm{aa}} = \lambda_{\mathrm{a}} - \gamma_{\mathrm{a}}\, \bar{\rho}_{\mathrm{aa}} - \left[i\,\hbar^{-1} \mathcal{V}_{\mathrm{ab}}\, \bar{\rho}_{\mathrm{ab}} + c.c. \right]$$

$$\dot{\bar{\rho}}_{\mathrm{bb}} = \lambda_{\mathrm{b}} - \gamma_{b}\, \bar{\rho}_{\mathrm{bb}} + \left[i\,\hbar^{-1} \mathcal{V}_{\mathrm{ab}}\, \bar{\rho}_{\mathrm{ab}} + c.c. \right]$$

$$\dot{\bar{\rho}}_{\mathrm{ab}} = -\left(i\,\omega_{\mathrm{ab}} + \gamma_{\mathrm{ab}}\right) \bar{\rho}_{\mathrm{ab}} + i\,\hbar^{-1} \mathcal{V}_{\mathrm{ab}} \left[\bar{\rho}_{\mathrm{aa}} - \bar{\rho}_{\mathrm{bb}} \right]$$

the **spontaneous emission factor** $n_e(\vec{\mathbf{k}})\, n_h(\vec{\mathbf{k}})$

and the so called ***absorption factor*** $1-\left[\frac{1}{n_e(\vec{\mathbf{k}})}-1\right]\left[\frac{1}{n_h(\vec{\mathbf{k}})}-1\right].$

In the spirit of the **free carrier model** and in the unsaturated limit the $n_\ell(\vec{\mathbf{k}})$'s are given by the *quasi equilibrium* Fermi-Dirac distributions -- *viz*.

$$n_\alpha(\vec{\mathbf{k}}) \qquad f_{\alpha\vec{\mathbf{k}}} = \left\{\exp\left[\beta\left(\varepsilon_{\alpha\vec{\mathbf{k}}}-\mu_\alpha\right)\right]+1\right\}^{-1} \qquad \text{[VII--46]}$$

Thus, the ***absorption factor*** is given by

$$1-\left[\frac{1}{n_e(\vec{\mathbf{k}})}-1\right]\left[\frac{1}{n_h(\vec{\mathbf{k}})}-1\right] = 1-\exp\left[\beta\left(\varepsilon_{e\vec{\mathbf{k}}}+\varepsilon_{h\vec{\mathbf{k}}}-\mu_e-\mu_h\right)\right]$$

$$= 1-\exp\left[\beta\left(\varepsilon_{\vec{\mathbf{k}}}-\Delta\mu\right)\right] \qquad \text{[VII--47]}$$

and the **spontaneous emission factor** is given by

$$n_e(\vec{\mathbf{k}})\, n_h(\vec{\mathbf{k}}) = \left\{\exp\left[-\beta\left(\varepsilon_{h\vec{\mathbf{k}}}-\mu_h\right)\right]\exp\left[\beta\left(\varepsilon_{\vec{\mathbf{k}}}-\Delta\mu\right)\right]+1\right\}^{-1}\left\{\exp\left[\beta\left(\varepsilon_{h\vec{\mathbf{k}}}-\mu_h\right)\right]+1\right\}^{-1} \qquad \text{[VII--48]}$$

where $\varepsilon_{\vec{\mathbf{k}}} = \dfrac{\hbar^2 k^2}{2m_r}$ and $\Delta\mu \equiv \mu_e+\mu_h = (E_F)_C-(E_F)_V-\varepsilon_g$. [45]

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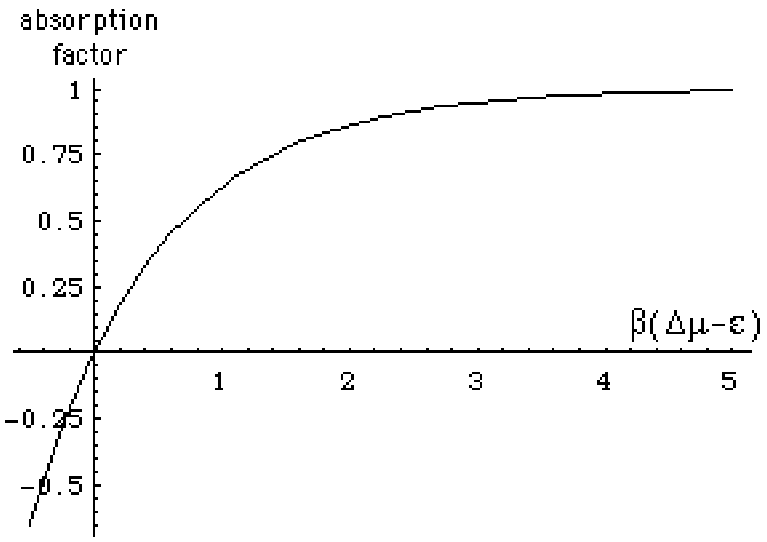
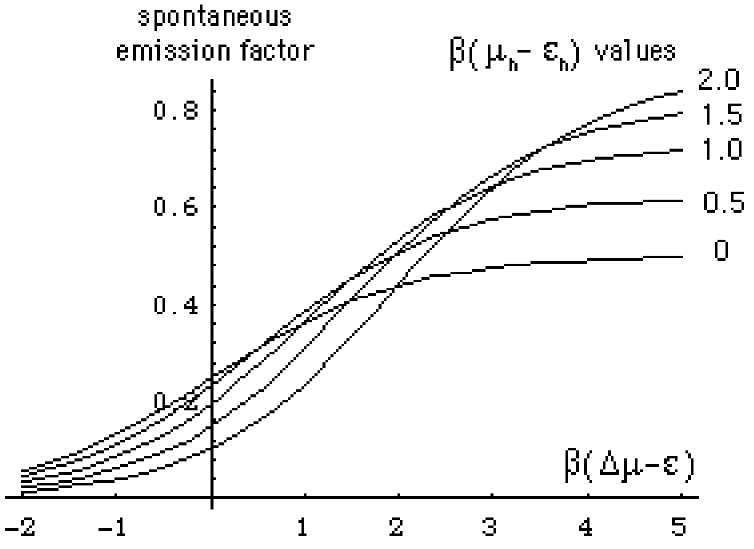
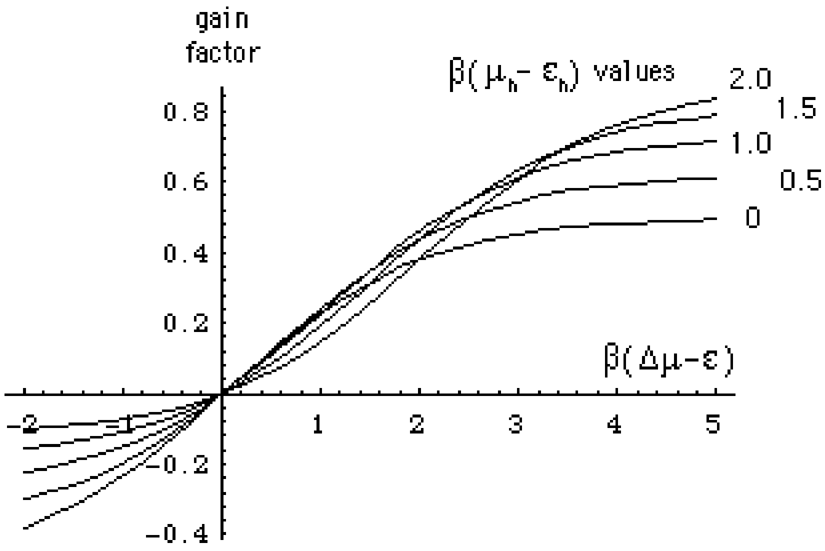
[45] For plotting purposes it is useful to write the complete **gain factor** as

$$\Delta(\vec{\mathbf{k}}) = \left\{\exp\left[\beta\varepsilon_g\left(\frac{x}{1+r}+\frac{r}{1+r}-x_F+x_h\right)\right]+1\right\}^{-1} - \left\{\exp\left[\beta\varepsilon_g\left(-\frac{r}{1+r}x+\frac{r}{1+r}+x_h\right)\right]+1\right\}^{-1}$$

where $r\equiv m_e/m_h$, $x=\hbar\omega_{\vec{\mathbf{k}}}/\varepsilon_g$, $x_F=\Delta E_F/\varepsilon_g$, and $x_h=\mu_h/\varepsilon_g$.







Therefore, **for population inversion** $\Delta\mu - \varepsilon = (E_F)_C - (E_F)_V - (\varepsilon_g + \varepsilon) > 0$ **which is a very stringent condition!**

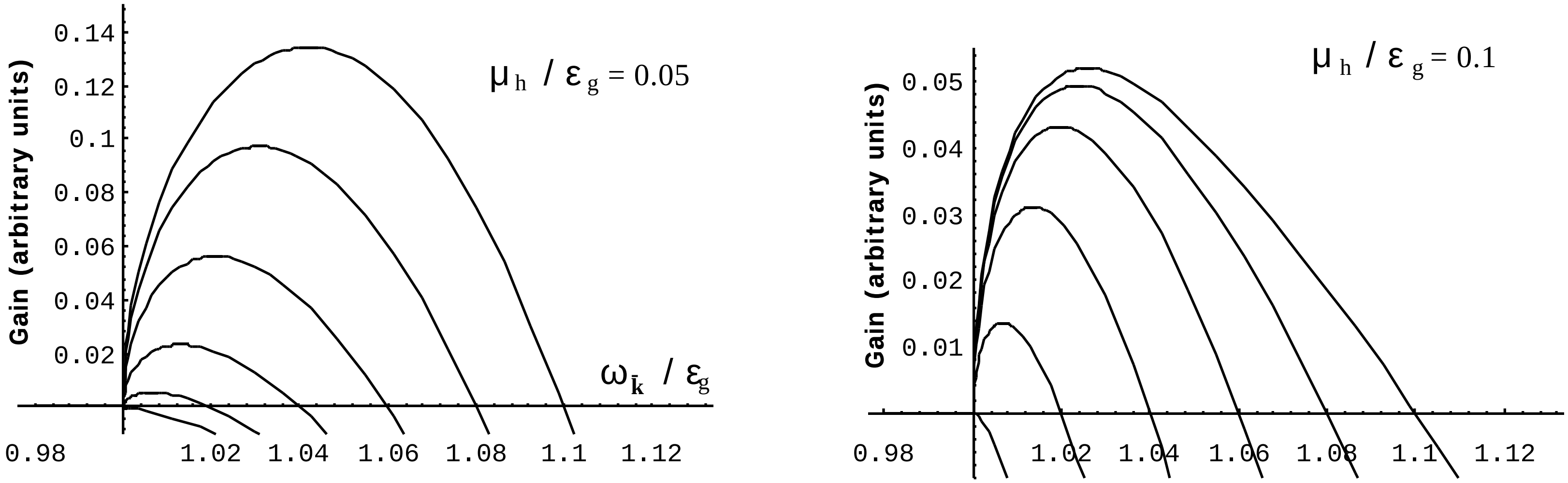
Adapting Equation [VI-26a] from earlier in this set of lecture notes,[46] we may see that

the **small signal gain** in a semiconductor, may be expressed in the form

$$Gain\left(\omega_{\vec{k}}\right) \quad \left|\overleftrightarrow{\mathbf{M}}_{v\,\vec{k},\,c\,\vec{k}}\right|^{-1} \Delta\left(\vec{\mathbf{k}}\right)\left\{\text{the density of } \textit{paired states} \text{ at } \vec{\mathbf{k}}\right\} \mathcal{L}\left(\omega_{\vec{k}} - \omega_n; \gamma_{ab}\right) \qquad [\text{ VII--49 }]$$

in bulk material the density of paired states varies as the square root of the energy.

Therefore, we may draw the following gain curves:



Gain in a bulk semiconductor with $\Delta E_F / \varepsilon_g$ = 1.00, 1.02, 1.04, 1.06, 1.08, and 1.10

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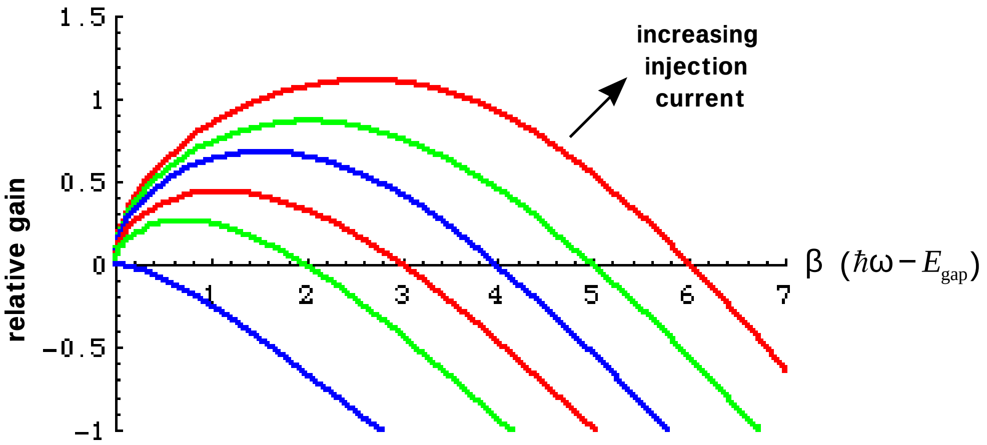
[46] That is $$\alpha_n = \frac{1}{2}\omega_n \frac{{}^{2}}{\hbar\varepsilon_o}\left(\frac{1}{\gamma_{ab}}\right)\mathcal{L}\left(\omega_{ab} - \omega_n; \gamma_{ab}\right) \bar{N} - \left(\frac{\omega_n}{2Q_n}\right)$$

GAIN VARIATION WITH CARRIER CONCENTRATION



QUANTUM CONFINEMENT

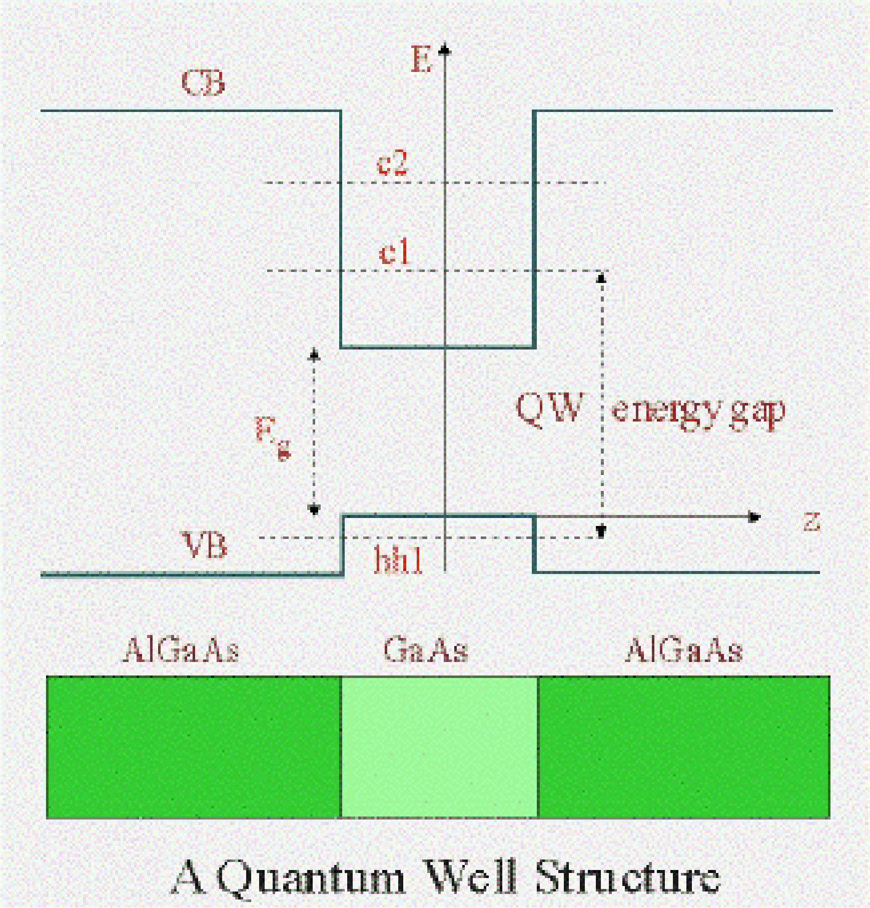
In an idealized quantum well

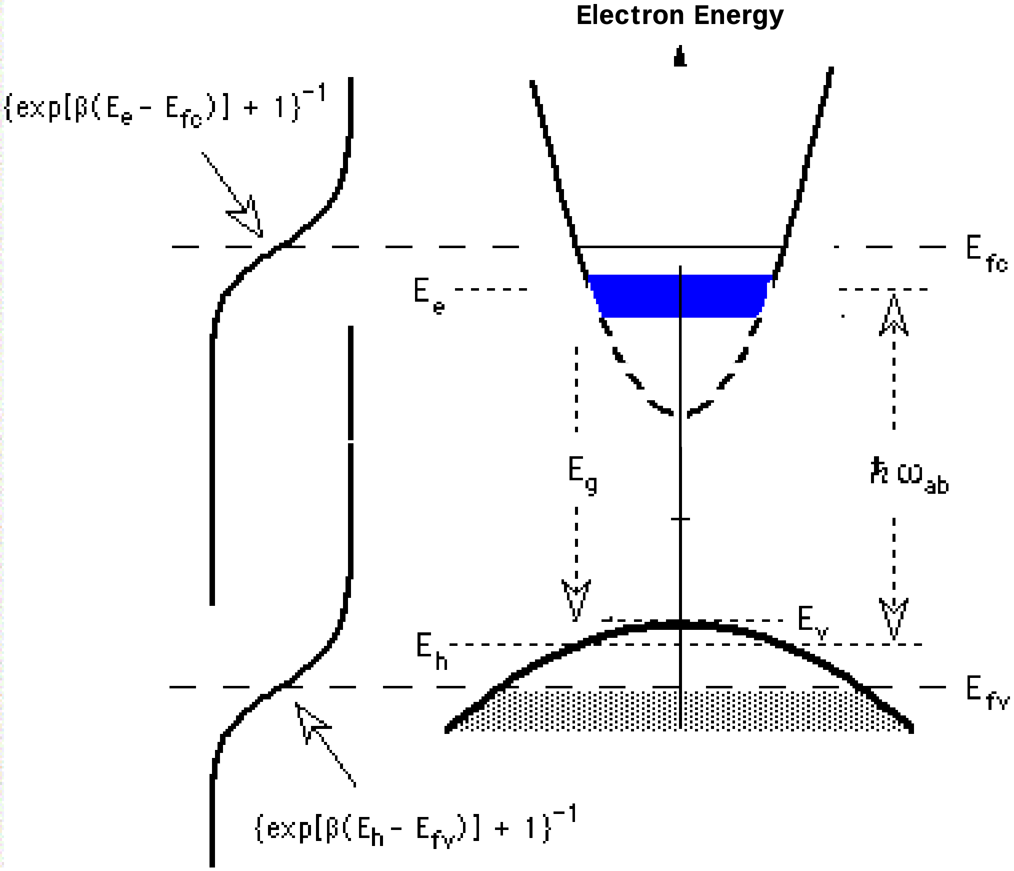
$$V(z) = \begin{cases} 0 & |z| < L_c/2 \\ \infty & |z| > L_c/2 \end{cases}$$

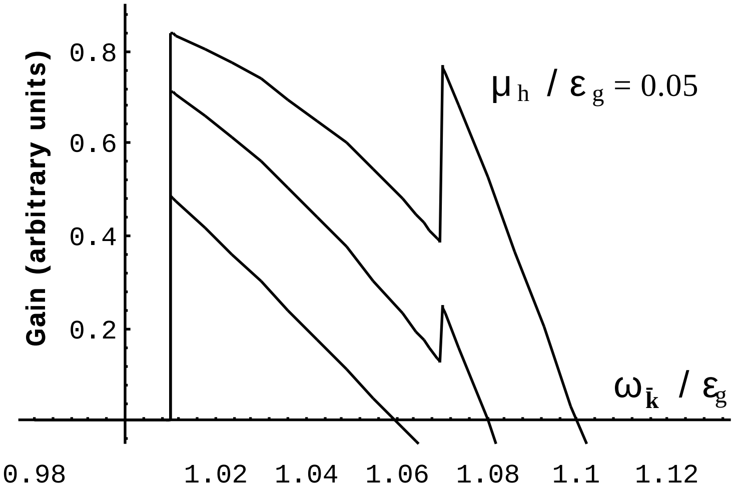
which yields a single electron energy

$$\mathrm{E}_n(\vec{\mathbf{k}}) = \frac{\hbar^2\, n^2\, \pi^2}{2\, m_z\, L_c^2} + \frac{\hbar^2\, |\vec{\mathbf{k}}|^2}{2\, m}$$

and density of paired states which is a constant or a staircase.


E
CB
c2
c1
E_g
QW energy gap
VB
hh1
z
AlGaAs
GaAs
AlGaAs
A Quantum Well Structure


Electron Energy
$\{\exp[\beta(E_e - E_{fc})] + 1\}^{-1}$
E_{fc}
E_e
E_g
$\hbar\omega_{ab}$
E_v
E_h
E_{fv}
$\{\exp[\beta(E_h - E_{fv})] + 1\}^{-1}$



Gain in a 2D semiconductor with $\Delta E_F / \varepsilon_g = 1.06, 1.08,$ and 1.10